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FINAL REPORT

Covering the Period

June 15 1948 to December 31 1953

Contract N8onr 60601

OFFICE OF NAVAL RESEARCH

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FINAL REPORT

Covering the Period June 15, 1948 to December 31, 1953

by

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Submitted in accordance
with the terms of the
contract noted above.

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SECTION I

Summary of the Work Described in the Previous Technical Reports

Submitted under the Terms of this Contract

1. INTRODUCTION

This is the Final Report to be submitted under Contract NS-cnr-60801, which was sponsored and supported at various times and in various amounts by the Office of Naval Research, Bureau of Ordnance, Bureau of Ships, Watson Laboratories, AMS, Air Force Cambridge Research Center, and the Atomic Energy Commission.

This contract was initiated and work started on 13 June 1948. It is now scheduled to expire on the present expiration date of 31 December 1953.

The primary purpose of this contract has been to sponsor research and development in the field of tunable narrow band optical filters. In its initial conception the aim was to develop and bring to practical realization the tunable filter conceived by Dr. B. E. Billings¹, which uses narrow polarization interference fringes arising from birefringent crystals to produce the narrow pass band (the Lyot² fixed filter), with the tuning being effected either by mechanical rotation of some of the polarizer or crystal elements or by means of the electrically variable retardation which can be provided by the electro-optic effect in crystals.

During the first two years of the contract almost all the emphasis was on the development of the fixed Lyot and tunable Billings

1. This conception was first described in a publication in the J. Opt. Soc. Am. 27, 738 (1947).

2. B. Lyot, Compt. rend. 197, 1595 (1955).

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filters to practical realization. In the case of the fixed filter the development proceeded to the point where the apparatus was susceptible to practical manufacture. This filter differed from Lyot's primarily in the use of ADP in place of quartz and calcite. A number of these fixed filters for the wavelength of H α have proven very useful for studies of the sun and its corona by astronomers. In the case of the tunable filter, although a number of research models were constructed, utilizing both mechanical and electrical tuning, its complexity and expense seemed an obstacle to its acceptance as a scientific tool for general use.

Towards the middle of 1950 there was somewhat of a shift in the emphasis of the work under the contract. Although continuing to work on tunable narrow band optical filters, the restriction to the polarization interference filter of the Lyot-Billet type was lifted. Various modifications of the Fabry-Perot interferometer and interference in thin evaporated films, metal or dielectric, were considered and developed, which might have considerable advantage in size, simplicity, and cost over spectrometers and monochromators of equal resolution. Fundamentally, this was the shortcoming of the original tunable filter. It was conceived as being a simple and inexpensive replacement for a large grating spectrograph, but it turned out to be about as costly and difficult to build.

In addition the work was made more practical by applying the tunable filter techniques to the scientifically and technically important field of isotope concentration measurement by spectrochemical techniques. Most of the practice was aimed at the hydrogen-deuterium problem as typical of the isotope measurement problem.

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The work under this contract has been reported in a series of Technical Reports which have been issued periodically. There are eight previous reports. It is felt that there is no advantage to reprinting for this final report the detailed material covered in these reports. However, in the next section brief abstracts and summaries of these reports will be given for reference purposes. Actually these interim technical reports were issued at appropriate times when it seemed possible to treat a particular technical subject in a fairly complete way, rather than to issue them over equal working times. An attempt was made to make each one an entity in itself.

Accordingly, after these brief summaries, the bulk of the text of this final report will deal with the developments under this contract since the last Technical Report was issued on 1 July 1953, and with any other material not previously covered in the Technical Reports. The interested reader is referred to the earlier Technical Reports for detailed information as to the work on the early phases of this project.

2. ABSTRACTS OF THE PRECEDING TECHNICAL REPORTS

All of these reports were issued under Contract N6-onr-60801, with the Office of Naval Research as the principal administrative agency.

2.1 B. E. Billings, "Summary of Tunable Filter Project", 30 December 1953.

This was a progress report which outlined the experimental programs then underway towards the development of fixed and tunable polarization filters, under the following divisions:

2.1.1 Procurement and manufacture of elements for a fixed Narrow Band Polarization Filter.

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2.1.2 Investigation of electrical tuning methods.

2.1.3 Development of mechanical tuning methods.

2.1.4 Improvement of the angular field of view of electrical tuning elements.

2.1.5 Development of fixed filters for isolating the spectral region to be covered by tuning.

2.1.6 Operation in the infrared.

2.2 W. M. Dravin and S. J. Sage, "Problem of Fixed Filter Development for Tuning Purposes", 11 March 1949.

One of the major problems in the development of a tunable filter is the manufacture of the basic fixed filter. A brief review of the theory of the Lyot type filter is described. The physical tolerances of the various elements is discussed, as are the techniques of orienting and optically finishing the crystal plates, temperature control and lamination.

2.3 W. M. Dravin, S. J. Sage and J. G. Anderson, Technical Report dated 11 August 1949.

This report covers the major activities undertaken during the period January 1 to August 1, 1949, in the development of a tunable narrow band optical filter. The problem of mechanical processing of elements is discussed with more detail than in the last report, most of the successful techniques having been determined at this time. Sections on birefringence variation with temperature, temperature control and housing, wave corrector plates and their measurements, tuning elements and additional uses for filter components have been included. Finally, a section covering problems for future investigation closes this report.

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2.4 B. J. Weichel, Technical Data, "The Index and Dispersion of Ammonium Dihydrogen Phosphate", 21 June 1950.

A compilation of the best values obtained from an evaluation of measurements at the National Bureau of Standards, University of Rochester, and Baird Associates, Inc. Final accuracy in index values is estimated as 5 in the sixth place beyond the decimal. Range 4000-6700 angstroms.

2.5 B. E. Billings, W. M. Drasin and S. J. Sage, Technical Report dated June 1950.

This report covers the entire development of the fixed and tunable filters from the inception of the contract in June 1948 until June 1950. It thus reports or summarizes most of the information in the preceding reports on fabrication of the fixed filter. In addition, the development of the tunable filter is treated in detail for the first time. Several tuning methods are presented, some of which were tried in the laboratory.

2.6 B. E. Billings, S. J. Sage and H. Zappod Jr., Technical Report dated 1 September 1952.

In this period the program was expanded by extensive development work on other types of narrow band optical filters. The use of multi-layer dielectric reflectors to obtain greatly decreased band pass and increased transmission in the solid Fabry-Perot interference filter is described. Filters 30 angstroms wide with peak transmission of 80% were produced. The simple inexpensive filters can be tuned over a range of at least 10% of the peak wavelength simply by tilting the filter. The

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use of these same multilayer reflectors to greatly improve the characteristics of the standard air-spaced Fabry-Perot interferometer is discussed. Application was made to the extension of Fabry-Perot interferometry into the infrared and to the high resolution study of isotope abundances. The tolerances on interferometer plate flatness is discussed, and apparatus for flatness testing was devised and described in the report. Mica cleavages silvered on both sides were developed as the best available solution to the problem of obtaining thick, flat, solid spacer layers for Fabry-Perot filters with band width the order of 1 angstrom. The difficulties of silver reflecting layers in the infrared is discussed. The mica plates were then coated with multilayer reflectors designed for about 1.7 microns. Experiments on the 1.7 micron doublet of mercury demonstrated that high resolution Fabry-Perot interferometry could be extended to the infrared, with tuning by tilting. The application of Fabry-Perot interferometry to isotope determinations is discussed, with application to the resolution of the mercury 2541 and the hydrogen 6563 isotope shifts and hyperfine structure. Finally, considerable further development on the tunable polarization interference filter is presented, with emphasis on extending the tuning range of the filter to cover very wide spectral regions. This requires low order retardation plates, and the available materials of low linear or circular birefringence are surveyed and evaluated.

2.7 B. E. Billings, D. E. Robinson, S. J. Sage and J. R. Welty, Technical Report dated December 1, 1952.

During the period covered by this report work centered on the application of the mica spacer Fabry-Perot interferometer to isotope analyses.

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A possible instrument for analyzing hydrogen-deuterium mixtures is discussed. This instrument is based upon the fact that the mica spacer can be adjusted to produce two oppositely polarized light beams, one transmitting at H₂ and the other at D₂.

Experiments have been performed to test the sensitivity and reproducibility of the system. These preliminary tests indicate that the instrument could be used successfully to measure the relative concentration of two isotopes.

2.8 R. O'B. Carpenter, S. J. Sage and N. Koppocf Jr., Technical Report dated July 1, 1955.

The Fabry-Perot interferometer with mica cleavages as the spacer, as described in the last report, is completely investigated in this report. The properties of mica seem to render it impractical for the usage envisaged, namely, the variability from sample to sample in birefringence, the impossibility of cleaving to a prescribed thickness, and its absorption.

An ADF birefringent polarization filter, a unique type of tunable filter, is described and analyzed completely in this report. It is found that it is suitable for only moderate accuracy isotopic analyses. Part of its difficulty arises from the fine structure and Doppler broadening of H₂ and D₂, and this effect is analyzed. The other difficulties are the high tolerances required in flatness of plates and in temperature control.

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3. PUBLISHED PAPERS AND TALKS

The following published papers and talks presented at scientific society meetings were supported by or stimulated by the work of this contract, at least in part.

3.1 B. H. Billings, "Tunable Narrow Passband Optical Filters", Winter Meeting of the Optical Society of America, February 20-22, 1947.

J. Opt. Soc. Am. 37, 521A (1947).

A polarization filter will be described which passes a region of the spectrum of the order of an angstrom in width and whose passband can be easily shifted to any wavelength in the visible spectrum. Previously, polarization filters have been designed which have a linear aperture of several centimeters and a transmission band an angstrom wide. The action of these filters depends on the superposition of the polarized channel spectra formed by X cut plates of quartz between parallel polarizers. Quartz plates were used whose thicknesses were in the ratios 1, 1/2, 1/4, etc., the alternate passbands of each plate were cancelled out by the black bands of the succeeding plate. Since the passband of such filters was fixed in wavelength, they had limited application. Interest in filters of this type was restricted mainly to astronomers. Analysis shows that the 1, 1/2, 1/4 retardation ratios in the fixed filter can be upset considerably without inducing any appreciable change in its optical characteristics. Tuning is accomplished by altering the retardation of the successive filter elements so that transmission maxima in the individual channel spectra coincide at the desired passband wavelength. This retardation change can be made mechanically for example by stretching supplemental

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plastic sheets in series with the elements, or can be made electrically by using Kerr cells or crystals with high electro-optic coefficients such as ammonium dihydrogen phosphate. The additional retardation never has to exceed a full wave. In the electrical case, a network could be arranged so that with a single control the passband could be shifted through the spectrum. A tunable filter will be demonstrated.

5.2 B. E. Billings, "A Tunable Narrow-Band Optical Filter", J. Opt. Soc.

Am. 37, 748 (1947).

Polarization interference filters are described which can have a pass band ranging from a fraction of an angstrom to several hundred angstroms in width. The pass band can be shifted to any desired region of the spectrum. These tunable filters are based on the fixed filters discussed by Lyot and Frans. The transmission band is formed by the superposition of the polarized channel spectra, produced by x-cut plates of quartz or other birefringent media placed between parallel polarizers. The birefringent plates have thicknesses in the ratio 1:2:4 etc. The tuning is accomplished by changing the retardation of successive elements so that transmission maxima in the various channel spectra coincide at the desired wavelength. The retardation change can be made mechanically, for example, by stretching supplemental plastic sheets in series with the filter elements, or can be made electrically by using Kerr cells or crystals with high electro-optic coefficients, such as ammonium dihydrogen phosphate. The additional retardation never has to exceed a full wave at the wavelength of peak transmission. The measured transmission of an experimental filter is shown. The electrical tuning method is particularly adapted to cathode

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ray oscillograph presentation of spectra. The filter also has possible application in color reproduction and colorimetry. With a pass band of a half-angstrom line of sight motion of solar prominences could be determined by the use of the Doppler shift of the prominence radiation.

3.3 to 3.6 B. E. Billings and R. O'B. Carpenter, "The Electro-Optic

Effect in Uniaxial Crystals of the Type $X \text{ H}_2\text{PO}_4$." J. Opt. Soc. Am.:

I. Theoretical, 39, 757 (1949)

When voltage is put across a crystal of the type $X \text{ H}_2\text{PO}_4$, the index ellipsoid in the crystal is rotated and the length of the axes changes. Two electro-optic coefficients are necessary to describe the effect in the most general plate of the crystal. The necessity for two coefficients is shown from consideration of the crystal symmetry. The behavior of Z-cut and X-cut plates is treated in detail. For X-cut plates with the voltage in the Z direction the crystal becomes biaxial with the plane of the optic axes at 45° to the X and Y crystallographic axis. The retardation along the normal to the plate is directly proportional to the voltage and independent of the thickness. The characteristics of the polarization interference pattern for excited X and Z plates are discussed.

II. Experimental, 39, 802 (1949)

In this paper it is shown experimentally that when a voltage is applied to the Z direction to a basal section of ammonium dihydrogen phosphate the crystal becomes biaxial. The normal to the plate is the bisector of the axes and the plane of the axes is inclined at 45° to the crystallographic axes. The location of the axial plane is independent of the magnitude of the voltage. The retardation along the normal is directly

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proportional to the voltage and is independent of the thickness of the basal section. Photographs are shown of the interference pattern obtained with polarized light when voltage is applied to the crystal.

The behavior of the crystal plate with an alternating voltage is also described. Between polarizers the crystal can operate as a light valve both for short pulses as in a camera shutter or for complex sound waves as in a sound-on-film device. It is shown that with circular polarized light 74 percent sinusoidal modulation introduces only 5 percent harmonic distortion.

III. Measurement of Coefficients, 40, 225 (1950)

The electro-optic coefficients of two crystals have been measured by both static and dynamic methods. For $\text{NH}_4\text{H}_2\text{PO}_4$, $r_{63} = 2.54$, $r_{41} = 6.25$; for KH_2PO_4 , $r_{63} = 3.15$, $r_{41} = 2.58$ (units 10^{-7} c.g.s., 20°C $\lambda 5560$). The variation of the electro-optic response with the frequency of the applied field is given and related to the piezoelectric vibrations of the crystal. A theoretical relation for the difference in the electro-optic constants measured at constant stress and at constant strain is derived, which provides a check on the consistency of the presently available electro-optic, piezoelectric, piezocaloric and elastic coefficients for these crystals. The use of a.c. methods to obtain high precision in polarization measurements is discussed.

IV. Angular Field of the Electro-Optic Shutter, 42, 12 (1952)

When voltage is applied to a basal section of uniaxial crystal of the type XH_2PO_4 , the crystal becomes biaxial. Between crossed polarizers it can be considered, then, to act as a light valve. The angular field

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of view is limited by the natural retardation of the crystal. This paper discusses methods by which this natural retardation can be minimized or effectively canceled so that the retardation with no voltage is essentially zero over a large angular field. In the first method a basal section of uniaxial crystal of opposite sign is placed in series with the slab to be excited. Another technique uses two slabs of similar crystal with a 90° optical rotator placed between them. Both these techniques are treated theoretically and experimental measurements are given to show the resulting angular polarization pattern. This pattern is shown both for the electrically excited case and the unexcited case.

3.7 H. O'B. Carpenter, "Induced Birefringence of AIP and K₂P as a Function of Temperature", Meeting of the Optical Society of America, Cleveland, October 28-29, 1950; J. Opt. Soc. Am. 40, 728A (1950).

In strongly paraelectric and ferroelectric crystals the photoelastic and electro-optic coefficients, along with the elastic, dielectric and piezoelectric properties³, exhibit large and anomalous temperature dependence⁴. Data have been obtained on the variation with temperature of the electro-optic r_{63} and the photoelastic q_{63} coefficients of AIP and KDP from room temperature down to their respective transition temperatures, i.e., in the para-electric region. Several techniques of making these measurements⁵ will be described and compared as to accuracy. The classical thermodynamic theory of the interaction of electrical and mechanical

3. W. P. Mason, Phys. Rev. 69, 173 (1948).

4. Zwicker and Scherrer, Helv. Phys. Acta 17, 346 (1944).

5. Carpenter, J. Opt. Soc. Am. 40, 225 (1950).

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influences within the crystal will be applied to this data in an attempt to relate the induced birefringence to a properly chosen set of macroscopic parameters in a temperature-independent fashion.

3.8 B. H. Billings, S. J. Sage and W. H. Renshaw, "A One-Wavelength Pass-Band Polarization Interference Filter", Winter Meeting of the Optical Society of America, New York, March 9-11, 1950; J. Opt. Soc. Am. 40, 882A (1950).

One of the main difficulties in the construction of a narrow band Lyot-~~Thom~~⁶ type polarization interference filter is the procurement of large slabs of transparent birefringent material. Previous filters have been built mainly of quartz. The narrower the band width, the thicker the required birefringent slab. A 1A width would need a quartz thickness of approximately 8.6 inches. Optical quartz of this thickness is essentially unavailable in quantity. A filter has been constructed using ammonium dihydrogen phosphate as the birefringent material. Although this crystal is easily available it is extremely difficult to work because of its softness and because it is water soluble. Furthermore, it has to be held to finer tolerances than the quartz slabs because of its higher birefringence. The filter which will be shown has mechanical tuning which enables it to be adjusted over a range of about 1A. It is planned in future work to tune⁷ these filters both electrically and mechanically over a wide spectral region.

6. B. Lyot, Compt. rend. 197, 1595 (1955); Y. Chama, Nature 181, 291 (1958).

7. B. H. Billings, J. Opt. Soc. Am. 37, 758-746 (1947)

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- 3.9 B. H. Billings, "A Birefringent Frustrated Total Reflection Filter", J. Opt. Soc. Am. 40, 471 (1950).

In the standard Turner Frustrated Total Reflection filter the pass bands occur in pairs which are oppositely polarized. The maximum theoretical transmission in any band is thus 50%. In this paper it is shown that if the high index layer of the filter is made of a birefringent material the oppositely polarized pass bands can be made to coincide so that the maximum theoretical transmission becomes 100% and the bands occur singly. Measurements are shown on a filter made with such a birefringent layer.

- 3.10 B. H. Billings, "Involuntary Polarization, and Depolarizers", Invited Paper, Winter Meeting of the Optical Society of America, Washington, March 1-3, 1951; J. Opt. Soc. Am. 41, 2347 (1951).

- 3.11 B. H. Billings, "Narrow Band Optical Interference Filters", Photographic Engineering 2, 45 (1951).

A publication of some of the principal results described in the more detailed technical report, reference 3.9.

- 3.12 B. H. Billings, "New Examples of Infrared Absorbers", (Thin Films in the Infrared), J. Phys. et radium, 407 (1950).

Thin films have found use in infrared light as optical filters, as bolometers and for reducing the amount of reflected light. A thin metal film can also be designed so as to wholly absorb a given wavelength in the infrared band.

Two types of filters are described. One is Dr. Turner's frustrated reflection filter, which transmits a narrow band in the visible

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spectrum. The present author has extended its use to infrared and to ultra-short waves of 1 cm wavelength. A sample is demonstrated, transmitting a band 0.1μ wide in the neighborhood of 3μ .

The second type is the Dennison Hadley reflecting filter, which reflects a set of infrared bands; several such filters are shown. The principle of the Dennison Hadley filter is the following: a thin metal film of 577 ohms/square resistance absorbs all the energy in a given wavelength, when placed a quarter wavelength in front of a perfectly reflecting mirror.

Such a thin film can constitute a bolometer, thus obviating the difficulty of having a satisfactory black body (all metallic blacks are grey in far infrared light): Such a black is no longer needed when the thin film is of appropriate resistance.

3.13 W. H. Dreisin, S. J. Sage, J. M. Anderson and J. G. Jelatis. "High Speed Photography with the Electro-Optic Shutter", presented at a meeting of the New England Section of the Optical Society of America, April 10, 1942.

The use of an artificially grown crystalline material, ammonium dihydrogen phosphate, as a high speed light valve is described. The orientation required to produce the optimum electro-optic effect, types of electrodes used, electronic requirements of the pulser unit activating the crystal, and the general performance of the shutter were included. Photographs of exploding firecrackers, exposed by the light of the explosions themselves, with exposure times controlled by the shutter and varying from 50 to 10 microseconds were shown. An actual demonstration

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SECTION II - A TECHNICAL REPORT

(This work was principally done in the period June 1 to December 31, 1955)

ABSTRACT

The Fabry-Perot interferometer is an extremely simple dispersing instrument requiring only two flat quartz plates with reflecting layers on one side held together by some form of mount to insure parallelism of the plates. In spite of its simplicity, it has long been recognized as the instrument of the highest spectral resolution available for the spectroscopy of close line structures. In this report its application to the problem of the spectrochemical analysis of hydrogen-deuterium mixtures is surveyed in detail.

The separation of the plates is determined to be 0.24 inches by the requirement that the deuterium fringes be exactly midway in the free spectral range between two successive hydrogen fringes. At such narrow spacings the instrument is not at all delicate or difficult to align, as compared to the more difficult problems arising in closer line structures, where the plates have to be spaced several inches apart.

In order to scan the wavelength spectrum, the interferometer is mounted in a vacuum tank. The fringes are observed through a circular aperture and photocell placed at the center of the well-known Fabry-Perot circular fringe pattern. By varying the air pressure in the tank, the fringes are collapsed, because of the varying index of refraction, and the wavelengths are scanned past the aperture. The resulting intensity versus wavelength is plotted on a strip chart recording microammeter. For air the scanning is only about .57 orders (or free spectral ranges) from

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vacuum to one atmosphere. This was improved by using SO_2 (.55 orders) and sulphur hexafluoride (1.5 orders).

The theory of the Fabry-Perot fringe shape is discussed. In any dispersing system the resolution and the contrast are the important parameters. The contrast is of considerable importance if deuterium in ordinary water (.015%) is to be detected, for the deuterium fringe is superposed on the hydrogen background.

After determining the theoretical factors which influence the resolution and contrast, an extensive experimental program was undertaken to achieve the maximum values of these two important parameters. The reflectivity of the reflecting layers, the flatness of the plates, and the width of the lines in the sources are the three limiting factors on resolution and contrast.

It is shown that multilayer reflecting layers are far superior to silver, and the multilayer prepared gave more than adequate reflectivity, resolution and contrast. The reflectivity was then less a limitation than the flatness of the best plates which could be obtained. A considerable discussion is given of the techniques of making multilayer reflectors, and of the apparatus which was constructed for the independent and precise measurement of the reflectivity.

It was necessary to considerably improve our apparatus for the independent determination of the flatness of the plates. Actually, in flatness determinations the same limits on reflectivity hold, so that it was necessary to first solve the reflectivity problem before the flatness of our excellent plates could be observed. When this was finally done,

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it was found that the plates were flat to somewhere between $1/50$ and $1/100$ of a fringe of red light.

At this point the width and contrast of our fringes were principally determined by the actual source width. A definite asymmetry in the H alpha line was observable, which is due to the Doppler broadened fine structure. The direction and magnitude of the asymmetry are in accordance with the structure, and the width is in accordance with a reasonable assumption as to the effective Doppler temperature in the source.

The recorder has to be somewhat improved in order to achieve the maximum signal to noise ratio. This involved adding additional input gain or sensitivity, and increasing the time constant by adding capacity to the input. Some care had also to be exercised to reduce the stray room light to a minimum, since any fluctuations in the strays would be disturbing and the deuterium light at such low concentrations is very weak. It is of course only the fluctuations in the dark current and instrumental background which limit the sensitivity of the method.

By means of all these experimental efforts, it was possible to observe the deuterium fringe in ordinary water, an effective contrast of about 5000 to 1.

A section in the report is devoted to a comparison of the light gathering power, or luminosity, of the Fabry-Perot with prisms, gratings and other dispersing elements. The high luminosity of this simple instrument gives it a further advantage over the other instruments. This is of particular importance when the application is energy limited. In this

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connection the multilayer reflectors are vital, for although silver can be evaporated to very high reflectivity, this is achieved at considerable expense in light transmission at the peak.

The report also discussed the problem of obtaining sources of high stability and narrow line widths. A mixing and discharging apparatus for hydrogen and deuterium gases was constructed. The most convenient source appears to be the water vapor discharge, and the final results were obtained with this source.

1. INTRODUCTION

For some months work under this contract has been concerned with isotope analysis by means of methods closely analogous to standard spectrochemical techniques. In particular, a number of simple interferometric methods have been investigated in the hope of devising instruments for separating closely spaced atomic lines originating from different isotopes of the same element which would be less bulky and expensive than large grating spectrographs.

In previous reports a polarization interferometer has been described which separates two lines in a simple two isotope problem into two beams polarized at right angles to each other. This led to an ingenious method where the wavelengths were not dispersed in ordinary space, but only in polarization space. Calculations were made to show how the system might work for hydrogen-deuterium analysis, as a simple, yet

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important, example of the two isotope problem. A fundamental difficulty in applying the method to low concentrations or with high accuracy arises in any case where the two lines had structure, which might arise from the fine structure of the lines or from the line broadening in the source. This difficulty arises in principle from the fact that in a sense there are only two polarization states, because any other state of polarization can be resolved into two components in the given states (usually taken at right angles). Therefore the polarization method, without additional dispersion in ordinary space, can only resolve two purely monochromatic lines.

Some work was also done on a mica Fabry-Perot interferometer system. This method was found to be impractical because of the great difficulty in selecting mica cleavage plates which had both birefringence value and thickness to the required tolerances.

The Fabry-Perot interferometer produces a series of very sharp interference fringes and produces a dispersing instrument of very high resolution and dispersion, but with a limited free spectral range because of overlapping orders. Therefore it is very highly suited to spectrum analysis problems involving only a very few very closely spaced spectrum lines, which is essentially the problem of isotopic analysis if all other lines in the spectrum can be eliminated with auxiliary filters.

Since mica is birefringent, i.e., has two indices of refraction, it produces two independent sets of Fabry-Perot fringes. Thus in the sense of the words used above, it is an instrument producing both dispersion and polarization. Since the polarization double beam method led

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to difficulties when there were more than two pure monochromatic components, reconsideration of the simple air spaced Fabry-Perot interferometer was undertaken.

One of the difficulties in the way of using an ordinary Fabry-Perot for spectrochemical analysis lies in the fact that two circular slits of different radii would be required to select two lines, and that precise tuning of the spacer thickness would have to be maintained in order to keep the two lines centered on the two slits. There is, however, a method which has been described in the literature⁸ for precise tuning of the fringes, which consists of varying the air pressure within the spacer layer. Since this varies the index of refraction of the spacer layer, the circular fringes are collapsed and expanded by this method. This led to the idea of a simple Fabry-Perot interferometer enclosed in a vacuum chamber, with the wavelengths passed by a single aperture controlled by varying the air pressure within the chamber.

In the next section this interferometer will be described in detail.

2. GENERAL DESCRIPTION OF THE FABRY-PEROT INTERFEROMETER

A Fabry-Perot interferometer consists of two highly reflecting layers separated by a spacing layer which maintains the two reflectors accurately plane and parallel. When air is the spacer the two plates are held apart by a three-legged device called a "spacer".

The theory and practice of the interferometer are described in detail in Tolansky's "High Resolution Spectroscopy". Only the most

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8. G. Defour, Thesis, Paris (1950).
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important equations of its behavior will be reproduced here for reference, and for considerations of the characteristics of the instrument which are critical for the E-D application.

Constructive interference occurs and bright sharp fringes are found at those wavelengths λ and angles θ (from the normal) satisfying

$$n\lambda = 2\mu t \cos \theta \quad (1)$$

where n is an integer, called the "order number" of the fringe, μ is the index of the spacer layer, t is its thickness.

A photograph of a typical set of circular fringes taken in monochromatic Hg 198 5461 Å light is shown in Figure 1.

Differentiating

$$\frac{dn}{d\lambda} = - \frac{2\mu t}{\lambda^2} \cos \theta \quad (2)$$

A parameter of interest is the so-called "free spectral range", the spacing in wavelength between successive fringes of the same wavelength, i.e., $d\lambda$ corresponding to $dn = 1$.

$$\text{Free spectral range } (\Delta\lambda)_f = - \frac{\lambda^2}{2\mu t \cos \theta} \approx \frac{\lambda^2}{2t} \quad (3)$$

The expression for the free spectral range in wave numbers is simpler, being independent of wave number

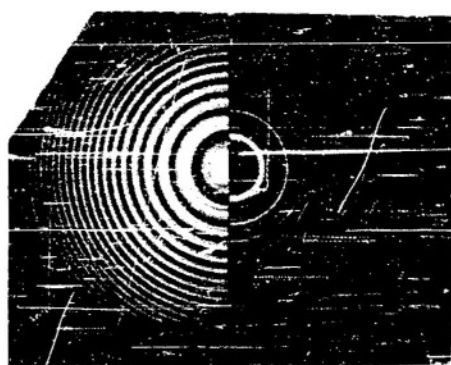
$$(\Delta\sigma)_f = \frac{1}{2\mu t \cos \theta} \approx \frac{1}{2t} \quad (4)$$

Now consider the application of the interferometer to hydrogen-deuterium analysis with H_{α} , where $\bar{\nu}_{H_{\alpha}} - \bar{\nu}_{D_{\alpha}} = 1.786$ angstroms. In order to



FIGURE '1

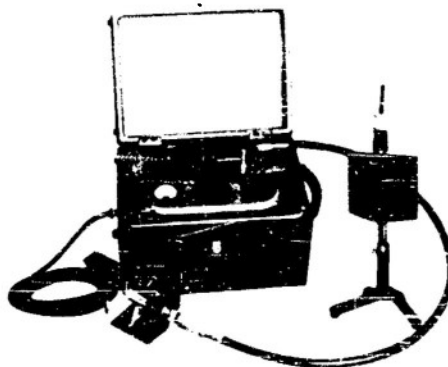
APPARATUS FOR HIGH DISPERSION INTERFEROMETRY MERCURY 198 SOURCE AND MICROWAVE EXCITER



ORDINARY
MERCURY

MERCURY 198

FRINGES TAKEN WITH FABRY-PEROT
INTERFEROMETER HAVING 43MM SPACER
DESCRIBED IN TECHNICAL CIRC. 502-1.



LAMP MOUNT & MICROWAVE EXCITER

The Mercury 198 source and exciter consists of two separate assemblies:

- a. The Mercury 198 electrodeless lamp containing approximately 0.2 mg. of Mercury 198 which has been prepared by transmutation of gold in a pile and contains no other isotopes. Argon is used as a buffer to enhance the mercury excitation. The lamp is 6 mm. in diameter by 14 cm long and is made of Vycor #7911 glass.
- b. The Mercury 198 Microwave Exciter operates at a wavelength of 12.2 cm in the 2400-2500 megacycle band. Radio frequency energy is generated in a continuous-wave magnetron oscillator and is carried in a flexible coaxial cable to the director which concentrates the external radiation on the lamp. The unit has a maximum power output of 125 watts. Relative power output is indicated by a per cent of maximum power meter on the front panel.

High-voltage direct current for the magnetron is provided by an AC-operated power supply which includes separate plate and filament transformers, two 816 rectifier tubes and a filter circuit. The input to the high-voltage transformer is controlled by a time delay relay, a timer and a Variac which controls the RF power output to the magnetron. The front panel Preheat and Power Switches control the application of filament and plate power respectively, while adjacent panel lights indicate switch positions. An AC operated motor-driven blower supplies cooling air throughout the cabinet.

The unit is portable and is packaged in a hand-rubbed quartered oak cabinet measuring 15" x 10" x 11". The exciter is designed for operation from a 60 cycle 115-volt AC power source.

The lamp mount has an aperture of $\frac{1}{2}$ " x $1\frac{1}{2}$ " and is furnished complete with pedestal and water jacket.

TECHNICAL CIRCULAR RD 502-3 Revised

Baird Associates, Inc.



PRECISION INSTRUMENTS FOR ANALYSIS AND CONTROL

33 UNIVERSITY ROAD, CAMBRIDGE 38, MASSACHUSETTS

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obtain the best possible separation, it is desirable to have the two wave lengths separated by exactly one-half order.

$$\Delta n = n_H - n_D = \frac{1}{2} = \mu t \left(\frac{1}{\lambda_H} - \frac{1}{\lambda_D} \right)$$

This determines the required spacing t between the two plates.

$$t = \frac{1}{4\mu \Delta \sigma} \quad (5)$$

where $\Delta \sigma$ is the difference in wave number for the two lines. For $H_\alpha - D_\alpha$, $\Delta \sigma = 4.145 \text{ cm}^{-1}$ and

$$t = .0805 \text{ cm} = .0237 \text{ inches} \quad (6)$$

There are of course other solutions separating the two fringes by $3/2$, $5/2$, etc. orders, and these require thickness 3, 5, etc. times as thick. However the tolerances on alignment, spacing, temperature control, etc., decrease with thickness, so the chosen solution is considered the simplest in practice.

Now the question arises as to how many fringes does the pattern shift from atmospheric pressure to vacuum. The change in order number is

$$n_{\text{air}} - n_{\text{vac}} = \frac{2(\mu - 1)t}{\lambda} \quad (7)$$

For air, $\mu - 1 = .00029$, so

$$n_{\text{air}} - n_{\text{vac}} = 0.55 \text{ fringes.}$$

Thus if the spacer thickness is adjusted to center the H-fringe exactly

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in the slit at atmospheric pressure, the D-fringe can be tuned through the slit by evacuating the chamber.

The maximum value of $\cos \theta$ and of the order number n occur at the center of the circular fringe pattern. For $t = .0803$ cm, n_0 is about 1837 waves. Integral orders from the normal occur at about $1^{\circ}49'$, $2^{\circ}41'$, $3^{\circ}15'$, etc.

The advantage of the pressure tuning method, of course, is that one may pick up the light photoelectrically from a circular aperture at the center of the pattern, and by varying the pressure in the instrument scan the spectral wavelengths past this aperture, recording the transmitted intensity versus pressure if desired.

Accordingly, a vacuum tank for holding the interferometer was designed and constructed. It was discovered that there was a scrap brass cylinder in the metal stores which came so close to the desired dimensions that a considerable saving in construction expense could be achieved by starting with this cylinder. The drawing for reworking this cylinder is shown in Schematic 48-2126. Figure 2 is a photograph of the finished tank. If it were not for using the available brass cylinder, the diameter could have been made somewhat smaller.

The simple aneroid type pressure gauge is an Ashcroft gauge manufactured by Manning, Maxwell and Moore. It presents atmospheric pressure to vacuum on a scale of 30 inches. The other fittings were an inlet for the vacuum pump and a small needle valve for admitting air. A simple Welch pump is adequate for this instrument. Holes were drilled in both the end plates and collimating lenses were sealed in with Apieson "Q"



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putty to serve as both window and lens. These are not shown in the figures, but were added later. The input condenser lens was of about 6 inches focal length, and is not required to be of particularly high quality. The focussing lens was a fine achromatic doublet of 100 cm focal length.

The interferometer is the standard Baird Associates, Inc. Fabry-Perot Interferometer. This instrument was originally designed similar to principles suggested by Mr. F. O. Westfall of the National Bureau of Standards. A photograph of the Baird instrument is shown in Figure 3.

The specified etalon spacing is .0237 inches. This is much smaller than any of the standard invar spacers furnished with the instrument. It was pointed out in Tolansky's book that in the manufacture of ordinary steel balls for bearings, the diameters are held to such close tolerances that these serve as the best practical spacers for Fabry-Perot work at very low separation. Our spacing was also somewhat smaller than is usually found in steel balls, but inquiries to the various steel ball manufacturers unearthed two balls close to the desired diameters. 1000 of each size were ordered: .0312 inches (.78 mm) and .0256 inches (.65 mm). The latter was used for most of the work.

A spacer plate had to be made to hold the balls from rolling around. The first one was made of .020 brass and is shown in Figure 4. Trouble was had with buckling, and better results were obtained by cutting a similar piece out of a mica cleavage.



Baird Associates
Better Analysis

FIGURE 3

APPARATUS FOR HIGH DISPERSION INTERFEROMETRY

FABRY—PEROT INTERFEROMETER

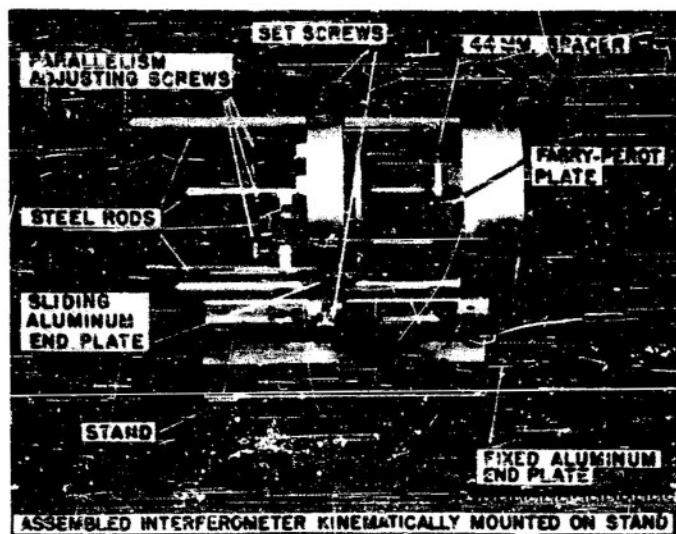
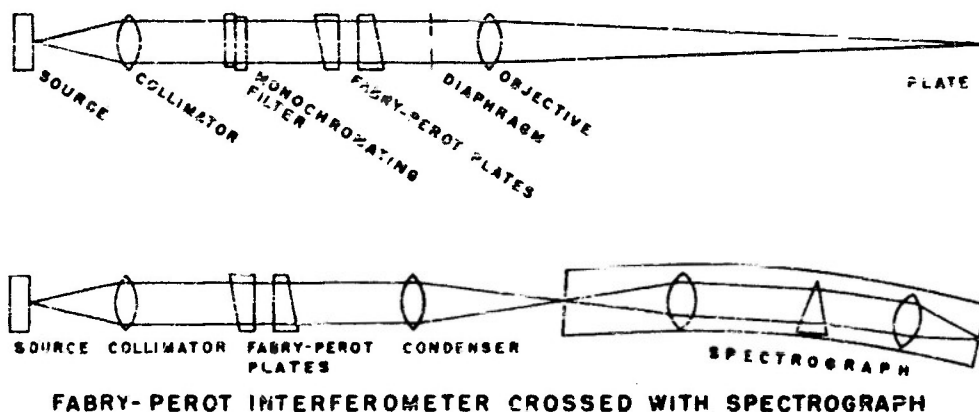


Figure 1

The theory of the Fabry-Perot interferometer is dealt with at length in the references given in the bibliography. Two illustrations of how it fits into an optical system are given below.

Features of the Baird Associates Fabry-Perot Interferometer are the remarkable ease of assembly, a minimum of adjustments, and a holder which accepts spacers from 0 to 100mm.



Baird Associates, Inc.



PRECISION INSTRUMENTS FOR
ANALYSIS AND CONTROL

33 UNIVERSITY ROAD, CAMBRIDGE 38, MASSACHUSETTS

Description of Instrument.

The Baird Associates Fabry-Perot Interferometer consists of two fused quartz discs with flat surfaces, spacers to hold the plates accurately parallel a fixed distance apart, and a holder to keep the plates lightly pressed against the spacer ends and to hold this assembly in an optical system.

The plates, Fig. 2F, are 2" in diameter and .625" thick. They are wedged 5 to 10 minutes of arc in order to displace the weak secondary pattern formed by the uncoated faces. One face of each of the two plates is figured to at least $\lambda/40$ measured at the mercury green line within a 40mm diameter circle concentric with the bevelled edge. The opposite side is figured to 1 wave. An arrow placed at the thickest part points toward the more highly figured face. The plates are supplied uncoated. For use a highly reflective, partially transmitting coating is deposited on the figured face. The coating is kept within a 34mm diameter circle so that the spacer ends rest on clear plate. The best performance is obtained with a silver coating. Baird Associates can supply silver-coated Fabry-Perot plates and will recoat plates on request (See price list). Silver coatings often have deteriorated substantially several weeks after deposition by the formation of sulfide. Because of this many users will wish to have coating facilities available locally.

The spacers, Fig. 2 C, D, and E, which hold the plates parallel a fixed distance apart are made of three invar rods press fitted into one or two stainless steel rings 2.75" in diameter. The spacer ends are optically finished to $1/10$ of a wave of mercury green light over an area at least 1 square millimeter.

When the polished spacer ends are in optical contact with the inner surfaces of the Fabry-Perot plates a yellow interference color is seen that permits easy checking of assembly. Standard spacer lengths are 7mm, 17mm, and 44mm. Other lengths up to 100mm are available on special order. The variation in spacer leg length is less than $\lambda/2$ of mercury green light.



Figure 2

The holder, Fig. 2B, which keeps the two plates lightly in contact with the ends of the spacer, consists of an aluminum base plate on which is mounted a fixed aluminum plate. Into this fixed end plate are press fitted three $1/2$ " steel rods. A second aluminum end plate slides on the three steel rods. One Fabry-Perot plate rests against 3 pins in a depression in the fixed end plate. The clear aperture of this end plate is a hole 1.375" in diameter. The other Fabry-Perot plate fits into the sliding end plate and is held in by three springs. The spacer fits inside the steel rods. Set screws clamp the sliding end plate in position and screws adjust the spring tension on the three spacer legs for the final parallelism adjustment.

The holder has a cone-slot-plane base (Kinematic mounting) for the precise repositioning on the ball bearing levelling screws of the stand. The outside dimensions of the interferometer are 6.75" x 4.875" x 4.375". The interferometer, stand, pair of Fabry-Perot plates, and 7mm, 17mm, and 44mm spacers fit into an attractive, compartmented oak case. Outside dimensions of case are 12.5" x 11" x 6.25". See photographs.

References

1. Tolansky, S. "High Resolution Spectroscopy" Menthuen and Co. Ltd. London 1947
2. Candler, C. "Modern Interferometers" Hilger and Watts Ltd. Hilger Division 1951
3. Meissner, Karl Wilh. Journal of the Optical Society of America Volume 31 (June 1941) pp 405-427 "Interference Spectroscopy Part I"
4. Williams, W. Ewart "Applications of Interferometry" E. P. Dutton and Company Inc. c. 1930

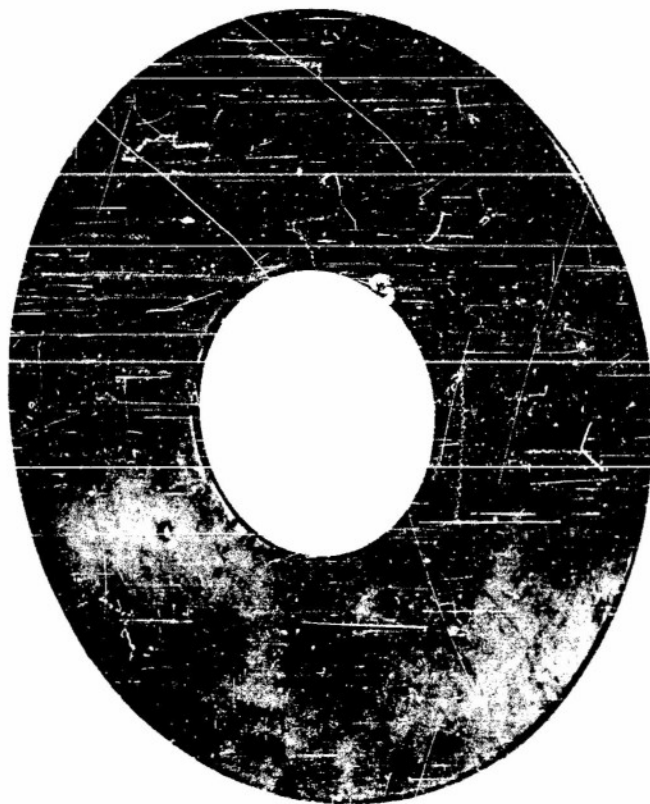


FIGURE 4

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The optical arrangement is shown in Figure 3. The fringes are in focus at the focal point of the 100 cm collimator. At this point a Geesco variable iris diaphragm was placed. A field lens and field stop gathered the light into the 1P21 photomultiplier. These served to eliminate all stray room light not coming from the immediate vicinity of the collimator lens. The photomultiplier power supply could supply various voltages stepwise. However after contrasts of several hundred were obtained, it became necessary to build a room light shield around the instrument, which was not in a dark room. A very satisfactory Leeds and Northrup Speedomax was available at the laboratories for recording the light as the pressure was varied in the tank. This instrument has standard scales of 2, 5, 10, 20, 50 and 100 microamperes full scale. A Ramson microammeter with 10 and 100 microampere scale was also occasionally used.

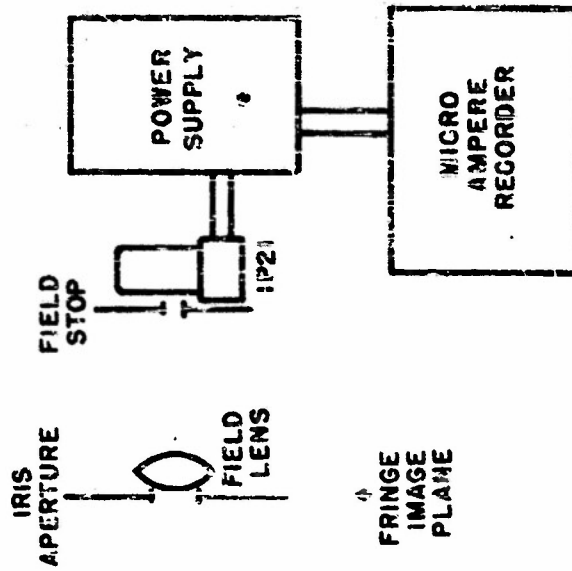
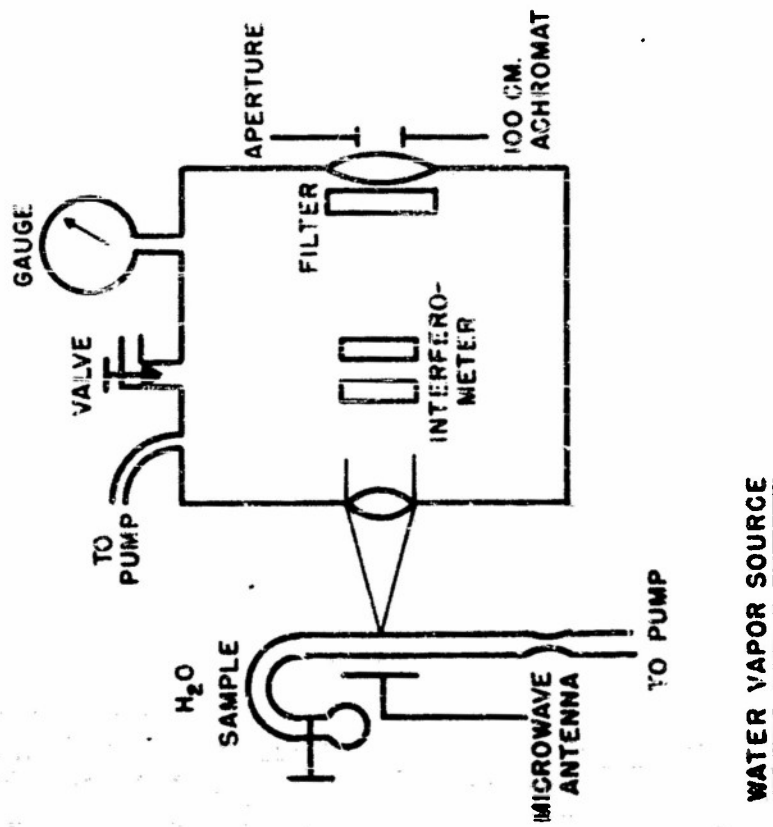
A refinement which has not been added to the apparatus as yet is a synchronization between the pressure in the tank and the recorder chart paper drive. This was done by Jacquinet and Dufour⁹ in a similar apparatus.

3. FRINGE INTENSITY DISTRIBUTION - CONTRAST, RESOLUTION AND TRANSMISSION OF THE FRINGES

Volansky gives the general expression as first derived by Airy for the fringe intensity distribution.

$$I = \frac{I^2}{(1-R)^2} \frac{1}{1 + \frac{4R}{(1-R)^2} \sin^2 \frac{\delta}{2}} \quad (8)$$

9. P. Jacquinet and G. Dufour, J. Recherches du C.N.R.S., Bellevue, 6, 1 (1948)



SCHEMATIC OF OPTICAL ARRANGEMENT
FABRY-PÉROT INTERFEROMETER WITH PRESSURE TUNING

FIGURE 5

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where $\delta = (2\pi/\lambda) 2t \cos \theta$.

A plot of this equation is shown in Figure 8, with the quantities of interest marked.

The ratio of the maximum transmission to the minimum is defined as the "contrast" of the fringes.

$$C = \frac{I_{\max}}{I_{\min}} = 1 + \frac{4R}{(1-R)^2} = \frac{(1+R)^2}{(1-R)^2} \quad (9)$$

This factor is important in determining the concentration accuracy of the instrument, or its sensitivity in measuring very low concentrations of deuterium, since the D light will be lost in the background of the H fringe. The minimum intensity occurs halfway between maxima, which is one of the principal reasons for choosing an etalon spacing making D appear one-half order from H.

Representative values of the contrast are:

<u>R</u>	<u>Contrast</u>	<u>I_{\min}/I_{\max}</u>
70%	31.0	3.22%
75%	49.0	2.04%
80%	81.3	1.23%
85%	152	0.66%
90%	360	0.28%
95%	1850	0.05%

It should be emphasized that the contrast depends only on the reflectivity R and not on the transmission or absorption in the reflecting layer.

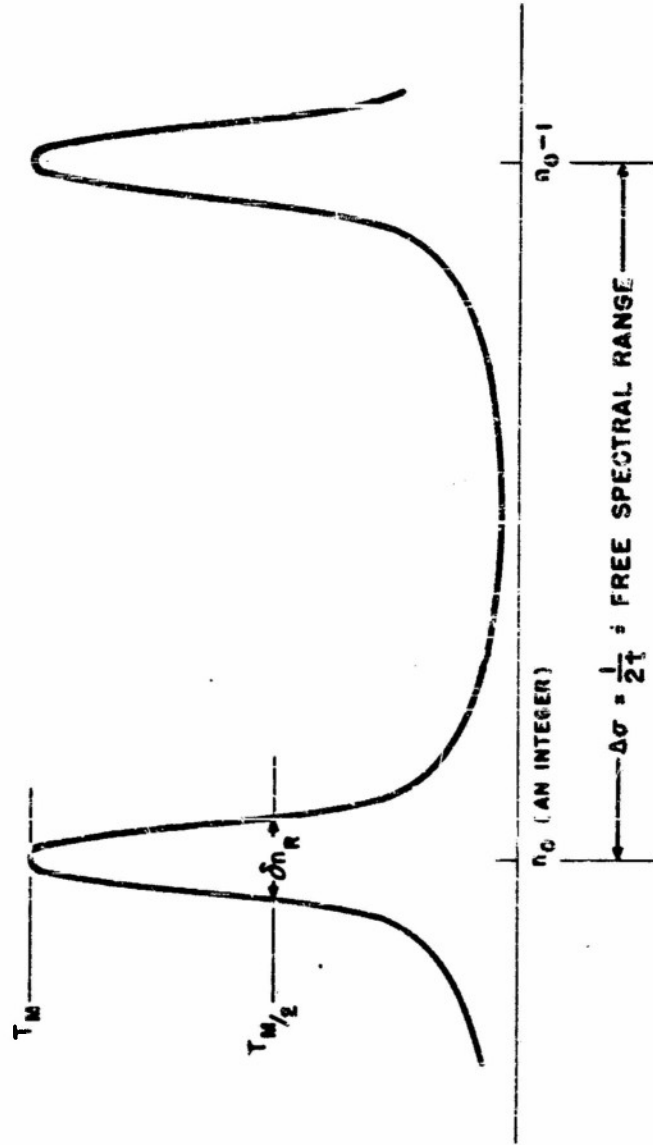
$$I(R, n) = T_M \frac{1}{1 + m \sin^2 \pi n}$$

$$m = \frac{4R}{(1-R)^2}$$

$$T_M = \left(\frac{1}{1-R} \right)^2$$

$$n = 2\mu\sigma t \cos \phi$$

$$\delta n_R = \frac{1-R}{\pi \sqrt{R}}$$



FRINGE INTENSITY DISTRIBUTION

FIGURE 6

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When we come to discuss the reduction of electronic noise the effect of the contrast on the resolving power in the intensity scale will be considered more carefully. It is the fluctuations in the background, not the background itself, that limits the accuracy and sensitivity in intensity measurement. In spectrum analysis one plots light intensity versus wavelength. The contrast has an important bearing on the resolution in the intensity scale. We now consider the resolution in the wavelength scale, which we will call by the limited term "resolution".

The wavelength resolution is limited by the half width of fringes which can be obtained. There have been a number of criteria or conventions used for the resolving limit of dispersing instruments, but they are all about the same. It seems most convenient to take as criterion that two lines can be just resolved if they are separated by total width between half maxima for either. This criterion is not greatly different from the well-known Rayleigh criterion.

From equation (6) for the fringe intensity distribution it is easy to show that the intensity is one-half when the order number $n = \frac{2\pi t \cos \theta}{\lambda}$ departs from an integer by

$$(\delta n)_{1/2} = \frac{1-R}{2\pi \sqrt{R}}$$

The total width between half maxima, measured in fractions of an order of interference, is just twice this, and will be designated by

$$\delta n = \frac{1-R}{\pi \sqrt{R}} \quad (10)$$

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This quantity is the resolving limit in orders of interference, and is a quantity depending only on reflectivity, not on wavelength, etalon spacing, nor angle of observation. Jacquinot and other French workers in the field have found it convenient to define the "finesse" F as the reciprocal of this resolving limit. This is a convenient parameter to be taken as the figure of merit of a Fabry-Perot interferometer.

$$F = \frac{1}{\Delta n} = \frac{\pi \sqrt{R}}{1-R} \quad (11)$$

The resolving limit in wavelength or wave number is now readily obtained by multiplying Δn by the free spectral range in wavelength or wave number.

$$\Delta \lambda = \Delta n \Delta \lambda = \frac{\Delta \lambda}{N} = \frac{\lambda \Delta n}{n_0} = \frac{\lambda}{2n_0} \approx \frac{\lambda^2}{2tF} \quad (12)$$

$$\Delta \sigma = \Delta n \Delta \sigma = \frac{\Delta \sigma}{N} \approx \frac{1}{2tF} = \frac{1}{2t} \Delta n = \frac{\sigma}{2n_0}$$

where n_0 = the order number.

The "resolution" as ordinarily defined in optical problems is now

$$\text{Resolution} = \frac{\lambda}{\Delta \lambda} = \frac{\sigma}{\Delta \sigma} = 2n_0 \quad (13)$$

The "finesse" is also a number of fundamental interest in the behavior of a Fabry-Perot interferometer, because it is approximately equal to the number of interfering beams of approximately equal intensity in the multiple beam interference. In other words, the beams of number greater than F have been diminished by successive reflection until they make a negligible contribution to the sum of interfering amplitudes. In

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the Lummer-Gehrke interferometer, for example, there are only a finite number of beams, but all have unity amplitude, since they are reflected at total internal reflection. A Lummer-Gehrke plate arranged to produce interference between just N beams has similar properties to a Fabry-Perot interferometer with finesse N .

The quantities now which depend solely on the reflectivity are the contrast C , the resolving limit Δn , the finesse N . These are plotted versus reflectivity in Figure 7.

In the case where R is .90 plus or minus .10, there is an approximate relation between C and N .

$$C \approx 0.4 N^2$$

4. TECHNIQUES OF MAKING REFLECTING LAYERS

The relative shape of the interference fringes in the Fabry-Perot depends only on the reflectivity of the layers. If there is absorption in the reflecting layer, as is always the case with thin metallic films, the transmission of the fringes is reduced everywhere by the same factor, the first term in equation (8). Thus for the peak transmission of the fringe, we can write

$$T_M = \frac{T^2}{(1-R)^2} = 1 - \frac{2A}{1-R} + \frac{A^2}{(1-R)^2} \quad (14)$$

since the absorption $A = 1 - R - T$.

This loss in transmission can of course become serious if one is operating in an energy limited situation. Such is very likely to be the case of one attempts to observe the deuterium alpha line from a

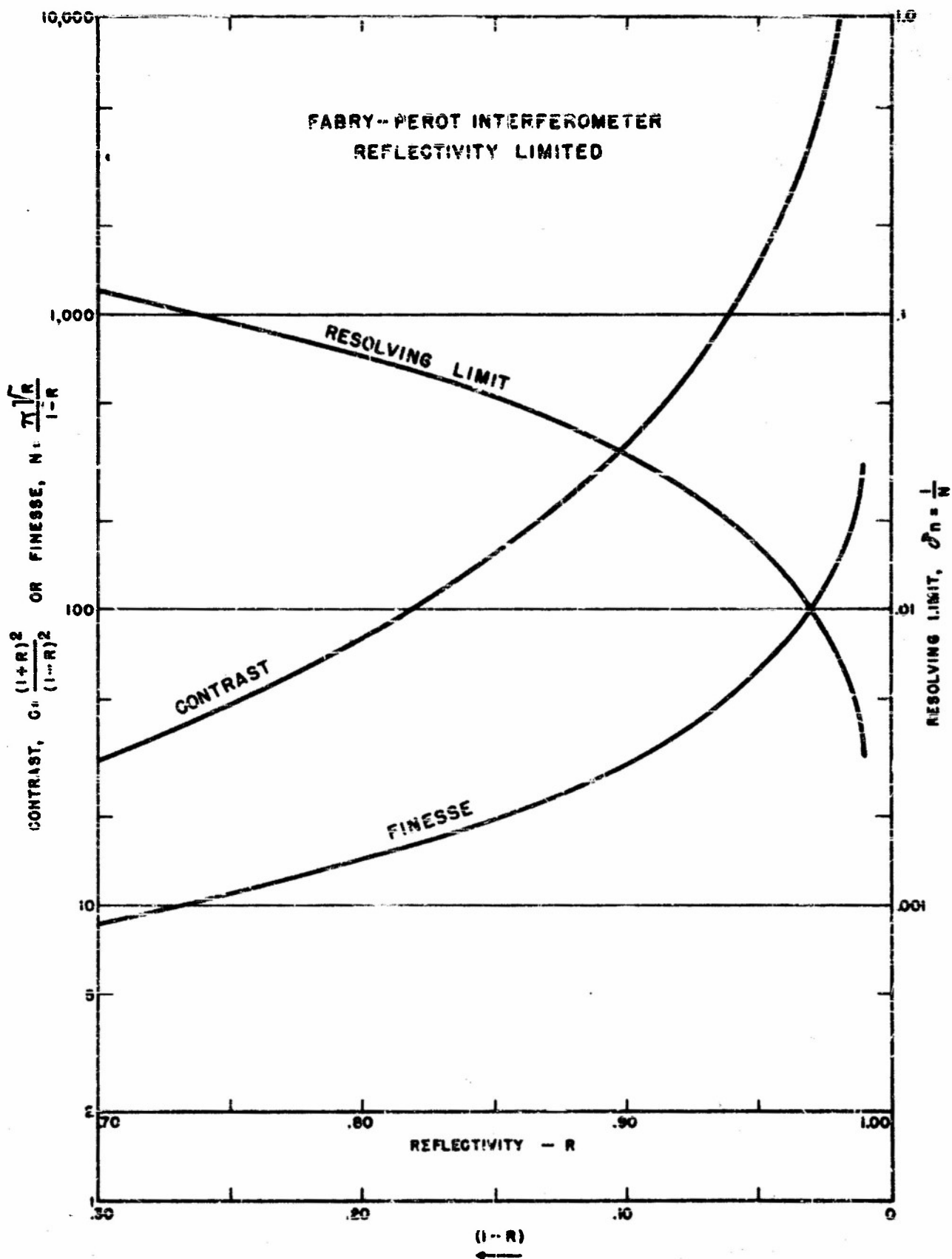


FIGURE 7

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sample with only a few hundredths percent deuterium. For the hydrogen discharge will be weak to start with if its line broadening is to be kept small, and photomultipliers have considerably reduced sensitivity at ~~5700 angstroms~~.

In Figure 5 is plotted the transmission T_{λ} of the interferometer for several fixed values of the absorption A . Following a suggestion of Jacquinot, the abscissa is the finesse F , which as discussed above, is ~~proportional to the resolution or to the square root of the contrast~~.

For some years it has been standard practice to make use of evaporated metal films for the reflecting layers, especially of silver and of aluminum. In this case, one cannot hold A to a fixed value while F is increased by increasing the thickness of the deposited layer. F , A , and hence T_{λ} are thus fixed functions of the thickness. This variation has been investigated or discussed by:

Fabry and Buisson, *J. de Physique* 2, 189 (1919). (For sputtered silver. These are obsolete, being inferior to evaporated silver.)

John Strong, *Procedures in Experimental Physics*; silver and aluminum in the red and blue.

Goos, *Zeits f. Phys.* 100, 95 (1936).

S. Tolansky, *Proc. Roy. Soc.*, 654 (1946); *Physica*, 649, December (1946); *High Resolution Spectroscopy*, Methuen and Company, London, 1947.

G. Dufour, *Thesis*, Paris, 1950.

The values the various investigators find for their practical evaporated layers are not always the same. This is partly due to the fact that the technique of evaporation, especially the purity of the metals,

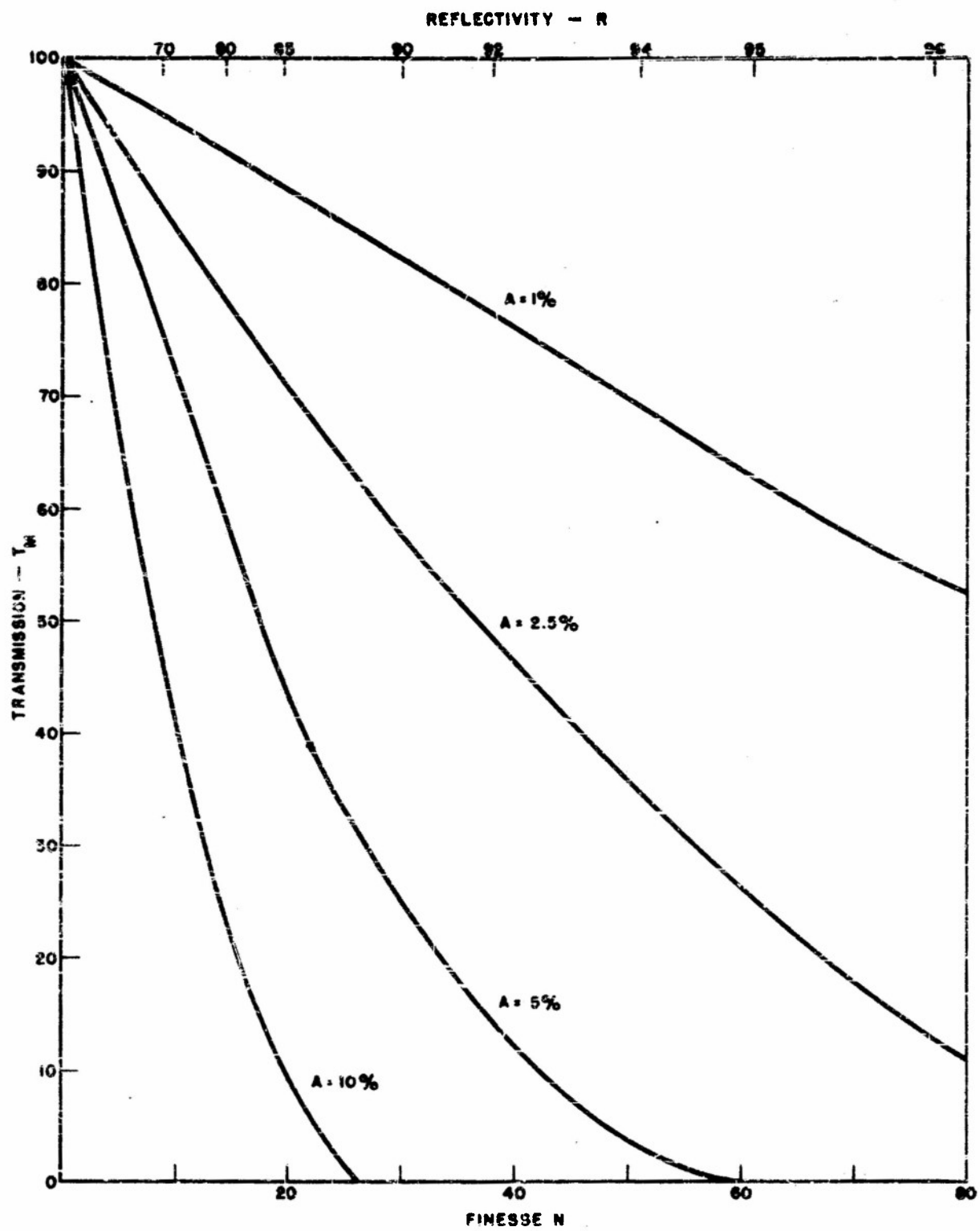


FIGURE 8

Beird Associates, Inc.

has a considerable effect. However there seems to be general agreement on the values in the region of sharply falling interferometer transmission. This is plotted in Figure 9.

It is further almost universally agreed that silver layers rapidly tarnish, suffering in both fineness and transmission. This can happen in a matter of days. One case measured accurately by Dufour is shown on the figure.

At wavelengths above 4000 angstroms silver is considerably superior to aluminum. However silver has a transparency around 3000 angstroms, so that aluminum is much better in this region. Aluminum in the red is shown in the figure.

In recent years a number of investigators have begun to make use of a new type of reflecting layer consisting of a series of layers of dielectric of alternating high and low refractive index. The history of this development is somewhat hard to trace, since a number of industrial, university and government laboratories all over the world have had extensive activity in the various applications of these multilayer dielectrics, and few bothered to publish much. One of the most complete studies with reference to these multilayers for Fabry-Perot interferometry is contained in Dufour's thesis cited above. The general theory of multilayer interference was published by Polster¹⁰. Beird Associates, Inc. has had considerable experience in this field, and some of this work is published in references cited above, and in a report on another contract.

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10. E. D. Polster, J. Opt. Soc. Am. 39, 1038 (1949).
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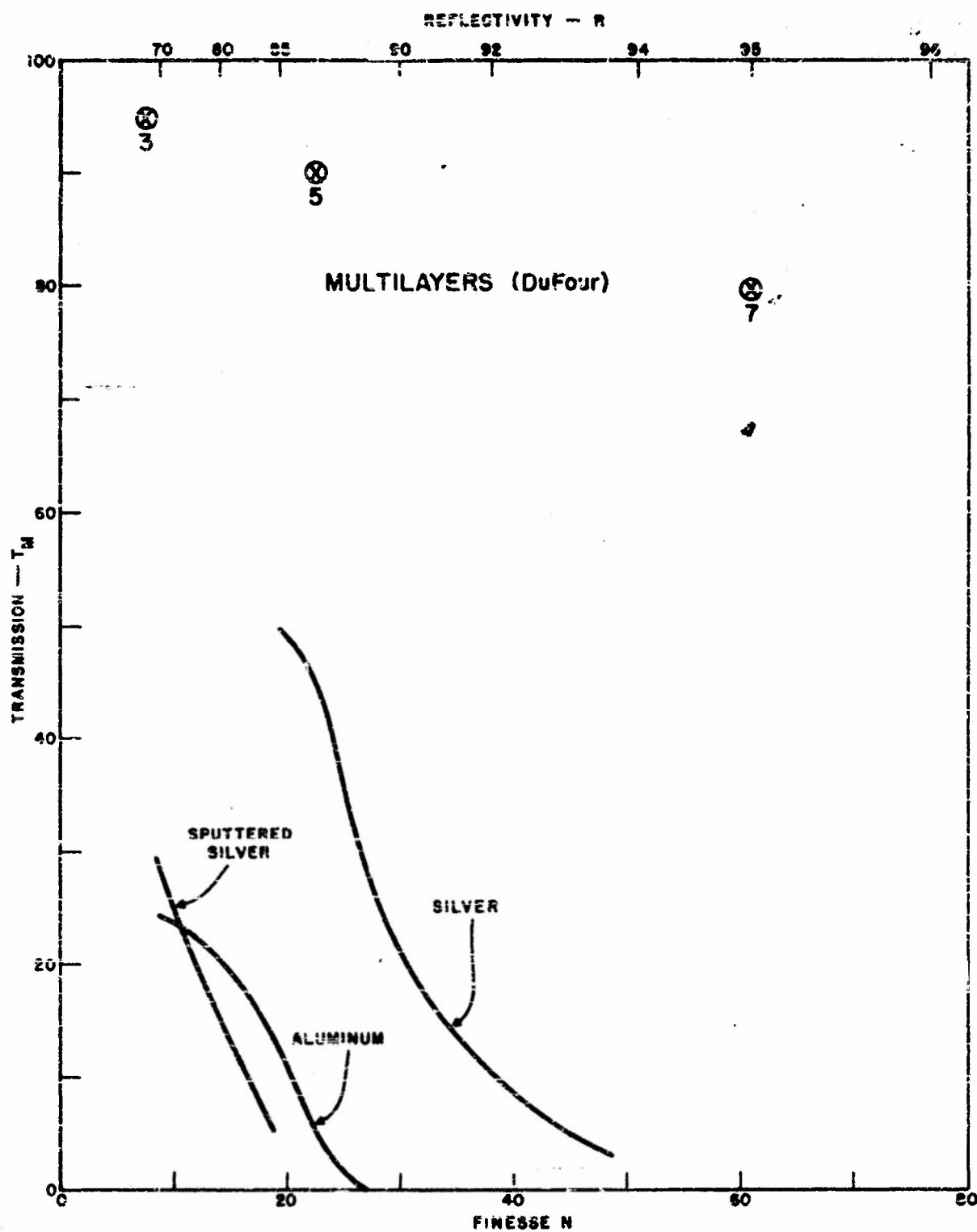


FIGURE 9

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Our first experiments with the pressure tuned interferometer were made with a pair of silver coated quartz plates which happened to be available. However, it was immediately recognized that the increased finesse and transmission which could be obtained with the multilayer reflectors would be vital to this difficult problem of resolving H and D light. We had ordered some especially flat quartz plates from Zeiss in Berlin for this job, and when they arrived, Messrs. E. Barr and S. Sullivan of our evaporating department initiated some experiments to perfect the technique of multilayer Fabry-Perot plates. The first experiments were on ordinary glass plates.

If one has an interface between a high and low refractive index, there will already be a considerable reflection described by Fresnel's equations. If further one stacks several of these layers together, each with an optical thickness of one quarter wave of light, the several reflecting beams will superpose in phase and the reflectivity very rapidly approaches unity. Of course a given thickness will be a quarter wave for only a single wavelength. Actually the general theory, checked by experience shows that the reflectivity is high and approximately constant for several hundred angstroms each side of the design peak. Approximate formulas for the peak reflection, and finesse when a pair is used in a Fabry-Perot, valid for any number of layers over 5, were suggested privately to the writer by Jacquinot. These are:

$$1 - R = \frac{4n_a n_g}{n_L^2} \left(\frac{n_L}{n_H} \right)^{Q+1} \quad (15)$$

where n_a , n_g , n_L and n_H are the indices for air, glass, the high and low

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dielectrics respectively, and q is the total number of layers, and

$$n = \frac{n_g \sqrt{R}}{1-R} \approx (1.7)^{q+1}$$

when $n_g = 1.52$, $n_L = 1.35$, $n_H = 2.5$.

Our best results were obtained with zinc sulphide and an equal mixture by molecular proportions of lithium, aluminum and sodium fluoride. This mixture appeared experimentally to give layers of improved stability and uniformity. Pure sodium fluoride is somewhat soluble in H_2O and is susceptible to haze. Cryolite, a mixture of 3 sodium to 1 aluminum, appears to be popular among some workers in the field. The best estimated indices from handbook values and from observation of the peak locations when the material is used as a spacer layer in a narrow band interference filter are:

1-1-1 mix	1.36
Cryolite	1.39
ZnS	2.38

These are averaged over the visible. The dispersion is not large in this region. Pure sodium and lithium fluorides were reported by Kohls¹¹ to have indices of 1.325 and 1.39 respectively.

Stone and Jenkins report that it is of considerable importance to purify ZnS by a pre-evaporation which presumably boils off any free sulfur. In deposition almost all workers determine the thickness by observing the rise and fall of transmission through a monitor plate inside the evaporating chamber as the thickness is increased. Apparently some

11. Kohls, Ann. d. Phys. 29, 433 (1957).

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monitor with a fresh surface for each layer, while others observed the total layers evaporated to this point. The writer has suggested that higher accuracy in this monitor determination might be achieved by using a monitor light at a different wavelength than that for which the multilayer is designed, where one could stop deposition at the maximum rate of change of transmission rather than on a broad peak. This has not been tried carefully as yet, however.

Applying the Jacquinot approximation to our assumed indices, we obtain the following values for the reflectivity to be expected for 5, 7 and 9.

<u>Number of Layers</u>	<u>Reflectivity</u>	<u>Finesse</u>
5	81.9	22
7	95.4	57
9	98.4	190

A five layer Fabry-Perot plate pair would be perhaps a factor of two more transparent than silver, but might be somewhat more difficult to make. The 7 and 9 layer reflectors are considerably superior to silver of the same reflectivity. The finesse has become so high here that the flatness of the plates will be the chief limitation to resolution and contrast. Actually it is not desirable to carry the multilayers too far, as it becomes increasingly difficult to maintain uniformity in thickness and good optical quality over so many layers. It was decided to go ahead and coat the Halle quartz with 9 layers for use in the interferometer.

It was soon apparent that it would be desirable to have accurate means for testing the reflectivity of Fabry-Perot plates in order to

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determine whether the theoretical expectations of the multilayer theory were being achieved. Also, it was of great importance to be able to measure reflectivity and flatness independently of each other, in order to determine which was the principal limitation on instrumental resolution and contrast.

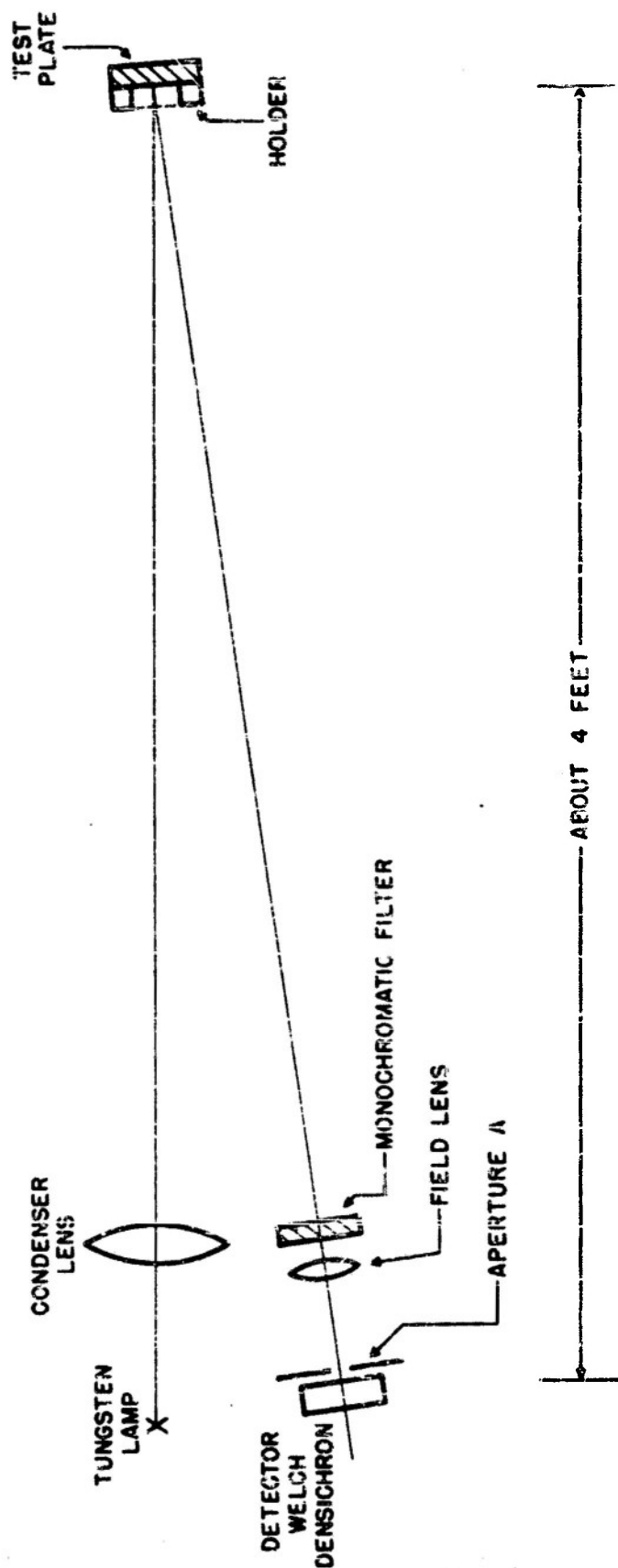
Our first brute force approach to this problem is shown in Figures 10 and 11. A tungsten lamp, narrow pass filter, Welch photometer head and mount for reflecting plates were built into a rigid system as shown. This enabled reflecting plates to be interchanged in the mount so reproducibly that the relative ratio of the reflectivities of two plates could be determined to about one percent accuracy.

Again Jacquinot in the course of his very fruitful visit to the laboratories suggested an improved apparatus. This was described by Giacomo¹². The instrument constructed on this project is shown in Figures 12 and 13. The procedure is as follows:

- a. Measure deflection with no plates equals K . (Usually the Welch Densichron is just set for zero density under this condition.)
- b. Then the deflection with the first plate will measure $KT_1(1 - R_d)$. R_d is just the correction for reflection at the air glass interface, about 4%.
- c. The second plate alone deflects $KT_2(1 - R_d)$.
- d. Then with both plates adjusted in Fabry-Perot parallelism, the deflection is

$$KT_1T_2 \frac{(1 - R_d)^2}{(1 - R)^2} \quad \text{where } R = \sqrt{R_1R_2}$$

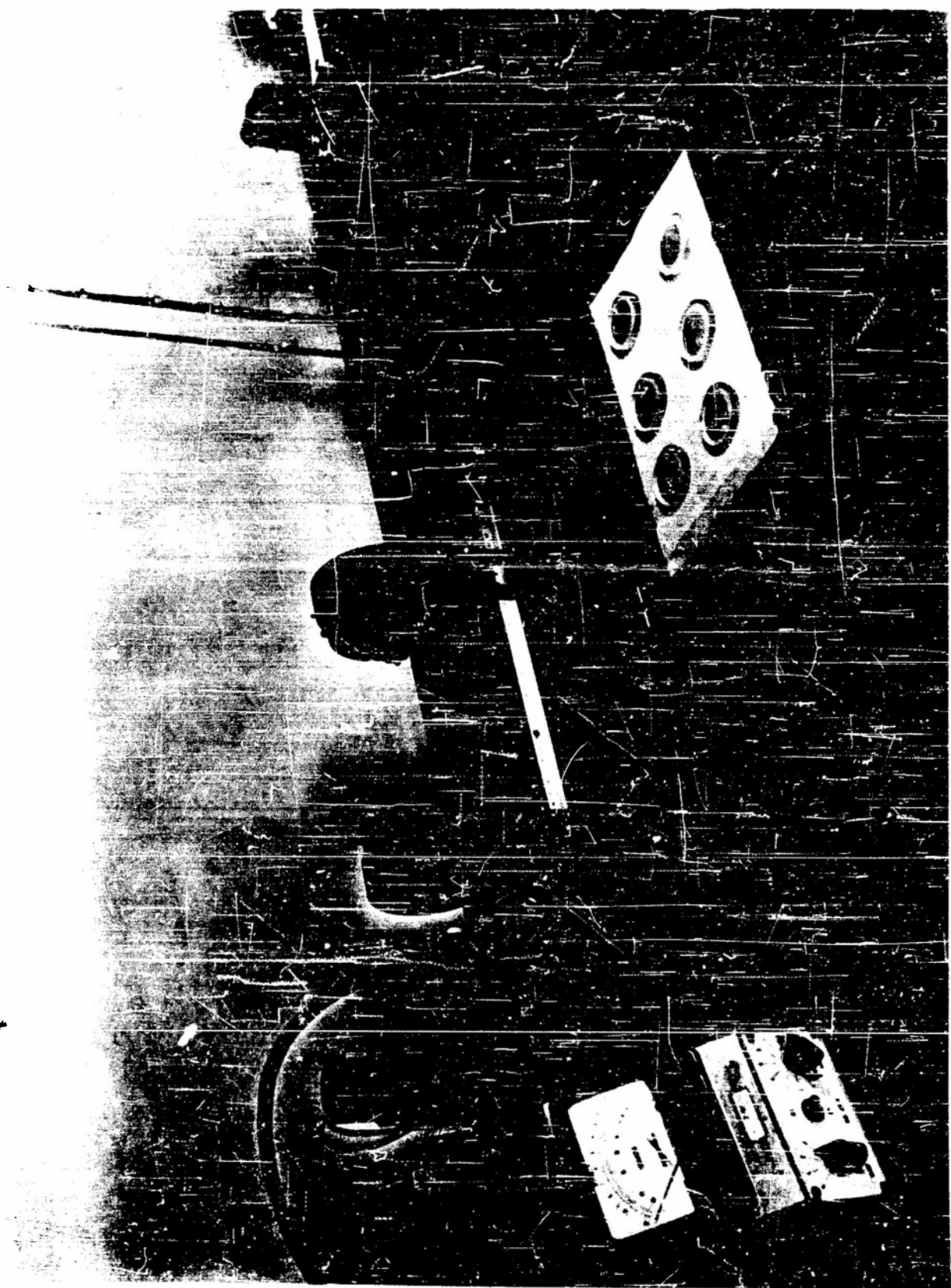
12. Pierre Giacomo, Compt. rend. December 1932, p. 1827.

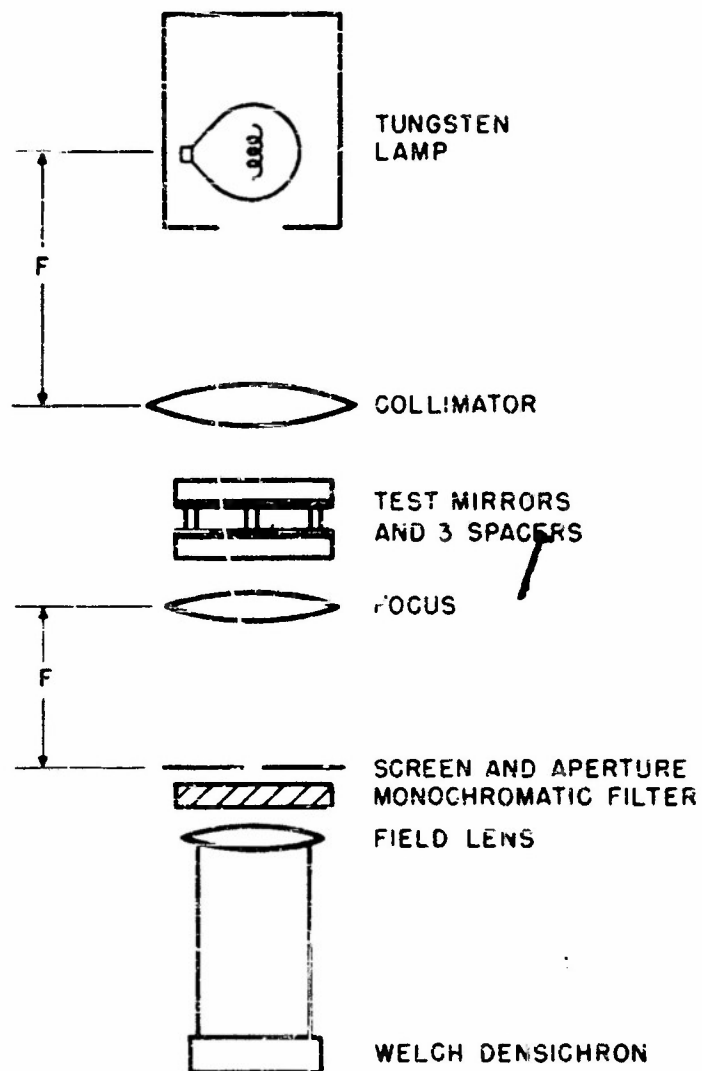


CONDENSER IMAGES SOURCE ON FIELD LENS.
FIELD LENS IMAGES APERTURE A ON TEST MIRROR.

APPARATUS FOR DIRECT REFLECTIVITY MEASUREMENT

FIGURE 10





APPARATUS FOR MEASURING I-R

FIGURE 12



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Thus solving,

$$T_1 = \frac{b}{a(1 - R_d)}$$

$$T_2 = \frac{c}{a(1 - R_d)}$$

$$1 - R^2 = \frac{bc}{ad}$$

or

$$R_1 R_2 = 1 - \frac{bc}{ad}$$

or if an approximate value for R is known,

$$1 - R = \frac{bc}{ad} \left(\frac{1}{1 + R} \right)$$

The advantage of this method over the former is that it directly measures $1-R$ rather than R . Although the other equipment could measure R to 1% accuracy, the accuracy in $1-R$ could become quite large, if R was, for example, about .95.

The result for the value d when measuring through both plates is obtained readily from equation (2) for the transmission of the Fabry-Perot. The condition is that the thickness of separation and the width of the wavelength band in the white source must be such that the source emission does not appreciably change over the free spectral range of the interferometer. Then one is justified in integrating the intensities of equation (9) over one whole order, obtaining the result given. The condition on the thickness e and the source width $\Delta\lambda$ is

$$e \gg \frac{\lambda^2}{2\Delta\lambda}$$

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For $\Delta\lambda = 30$ angstroms at R_x , this prescribes $\phi > 25^\circ$.

Adequate spacers were found by cutting small chips out of some celluloid material. The instrument was arranged vertically so that one plate could be laid down, spacers on it, and the second simply laid over the spacers. It is not at all difficult to determine whether the two plates are sufficiently parallel. A small aperture just larger than the filament image is laid on the stage and moved until the image completely passes through it. If there is a wedge in the plates, multiple images will be observed. When the plates are parallel, all these images will pass through the aperture, which is all that is required.

Although the method only gives the product of $R_1 R_2$, with three plates all three R 's can be determined absolutely by measuring the products for all three pairs.

By means of this apparatus, the reflectivities of our 5, 7 and 9 layer plates were measured to be 87, 95 and 97 percent respectively, values in good agreement with the theoretical prediction.

Some of the experimental multilayer plates are shown in the photograph near the instrument for determining reflectivity.

The resolution and contrast of the Muller quartz plates, as limited strictly by the reflectivity, were considerably greater than was actually obtained in the Fabry-Perot fringes in R_x light. It was thus evident that the effect of plate flatness and source line width would have to be investigated.

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3. EFFECT OF FLATNESS, SOURCE WIDTH AND APERTURE SIZE

So far the consideration of the relative shape, resolution and contrast of the fringes has been limited to the effect of the reflectivity of the plates. It has been assumed that the plate thickness t and the wavelength λ were quite constant. When these ideal conditions are not fulfilled, the distribution of plate flatness defects and of wavelengths in the source may contribute to fringe broadening and loss of contrast. The effect of the size of the circular aperture before the photocell is also to be considered. It is of interest to obtain the expression for the absolute light energy transmitted through the entire instrument, both at the peak and at the minimum of the fringes, for noise considerations.

In any optical system the total light transmitted is given by the product of source brightness by the area and the angular aperture of the system, multiplied by any transmission factor. The exact expression for the absolute energy transmitted by the Fabry-Perot instrument considered here is thus given by

$$E = \int \int \int J_{\sigma} d\sigma \cdot A(t) dt \cdot 2\pi r dr \cdot I(R, \sigma, t, \theta) \quad (16)$$

This triple integral is derived in the following way. J_{σ} is the spectral brightness distribution of the source. $J_{\sigma} d\sigma$ is thus the total brightness in the range $d\sigma$. It is convenient to use wave number rather than wavelength as the variable because the expression for order number n is linear in wave number, but inverse with wavelength. $A(t)dr$ is the area of the Fabry-Perot plates having thickness between t and $t+dt$. The distribution function $A(t)$ thus describes the departure of the plates from

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exact flatness and parallelism. $2\pi\phi$ is the angular aperture of the light as it passes through the interferometer. That is, it is the solid angle subtended at the plates by the annular ring in the image plane of the fringes between θ and $\theta+d\theta$. In practice an aperture is placed in this plane and the light passed through this aperture to a photocell. Consideration is limited to centered annular rings in order to have to deal with only a one dimensional integral for the solid angular aperture. In the experiments the aperture was always a simple circular opening, but the results are equally simple and general for any annular ring extending from θ_1 to θ_2 , the one dimensional integral in θ being taken between these limits. The final factor I is the transmission of the interferometer, which has been previously given in equation (8), but which is now rewritten in terms of the variables n and σ .

$$I(R, \sigma, t, \theta) = T_n \frac{1}{1 + n \sin^2 \left(\frac{\sigma}{2} \right)}$$

where T_n is nearly one for multilayer reflectors (no absorption),

$$n = \frac{4R}{(1 - R)^2}$$

$$n = 2\pi\sigma \cos\theta \approx 2\pi\sigma(1 - \frac{1}{2}\theta^2) \quad \theta \ll 1$$

There are four more or less sharply peaked distributions in this triple integral, the wavelength distribution, the flatness distribution, the reflectivity limited fringe shape and the aperture transmission, which can be considered as an ideal square wave distribution in θ . The width of each of these distributions makes its contribution to the overall fringe

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width. It is the purpose of this section to consider with some rigor these relations.

The source shape is determined by any fine line structure and by the Doppler broadening. A few remarks on the flatness distribution seem to be in order. There are certain simple types of thickness defect which lead to simple analytical distributions which can be easily treated. First, there may be random roughness of the plates on a rather small microscopic scale, due to the roughness of the polishing process. It seems reasonable to assume that these defects have a Gaussian distribution. A second simple type is a wedge, i.e., a departure from parallelism. If the aperture which is utilized is square, it is obvious that the distribution of thickness in the wedge is a square wave. These two types of distribution are indicated graphically in Figure 14.

It is perhaps surprising to find that the distribution for a centered spherical error, which is a common flatness error, when used with a circular aperture, is likewise a square wave, thus identical in shape to the previous case.

It is of considerable convenience to make a change of the variables of integration in the general formula (16), so that thickness, wave number and angle θ will be reduced to the same dimensions and can be considered on a common level. The order number n of the interference is an ideal variable for this purpose. We write

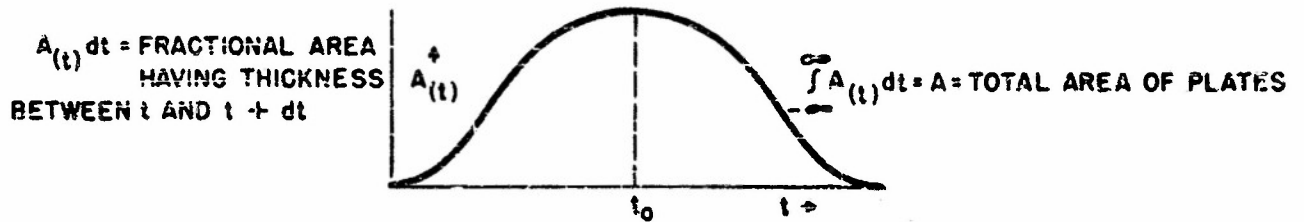
$$t = t_0 + t'$$

$$\delta = \delta_0 + \delta' \quad (17)$$

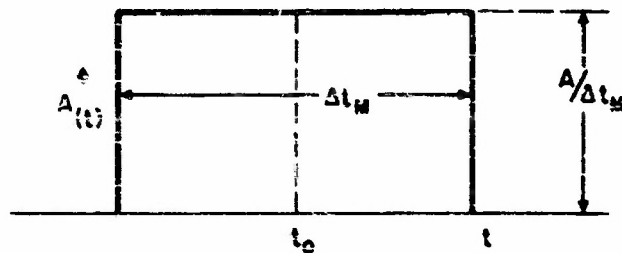
$$\theta = \theta_0 + \theta'$$

FIGURE 14
DISTRIBUTIONS OF SURFACE DEFECTS

1. GAUSSIAN DISTRIBUTION OF MICROSCOPIC ROUGHNESS



2. WEDGE OR SPHERICAL DEFECT



a. WEDGE



b. LENS DEFECT



AREA OF PLATE HAVING THICKNESS BETWEEN y AND $y + dy = 2\pi x dx = 2\pi R dy$

FOR SINCE $y = \frac{x^2}{2R}$, $R dy = x dx$

R = RADIUS OF CURVATURE

NORMALIZATION: $A = \pi x_m^2 = 2\pi R \Delta t_m$

$$A_{(t)} = \frac{A}{\Delta t_m}$$

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where $t_0 = \bar{t}_0$ and θ_0 are the average values of their respective distributions, and the primed variables are the small deviations. Then the expression for n , to the first order in the small deviations, becomes

$$n = 2t_0(t_0 + t')(\theta_0 + \theta') \left\{ 1 - \frac{1}{2}(\theta_0 + \theta')^2 \right\}$$

$$n = \left\{ 2t_0\theta_0 \left(1 - \frac{1}{2}\theta_0^2 \right) \right\} + 2t_0\theta_0 t' + 2t_0\theta_0 \theta' - 2t_0\theta_0 \theta_0 \theta'$$

which we write as

$$n = n_0 + n_t + n_\theta - n_a$$

introducing the variables n as the equivalent order numbers of the various deviations from n . That is

$$\begin{aligned} n_0 &= 2t_0\theta_0 \left(1 - \frac{1}{2}\theta_0^2 \right) \\ n_t &= 2t_0\theta_0 t' \\ n_\theta &= 2t_0\theta_0 \theta' \\ n_a &= 2t_0\theta_0 t_0 \theta_0 \theta' \end{aligned} \quad (18)$$

This change of variables is then introduced in equation (16), giving

$$F = \frac{2\pi}{n_0} \int_{n_t} \int_{n_\theta} \int_{n_a} J(n_\theta) dn_\theta \cdot A(n_t) dn_t \cdot dn_a \cdot I(R, n) \quad (19)$$

It is assumed that the distributions J and A have been re-expressed with n as variable, maintaining $J(\theta)d\theta = J(n_\theta)dn_\theta$ and

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$A(t)dt = A(n_t)dn_t$. This requires

$$J(n_t) = \frac{1}{n_t t_0} J(t)$$

$$A(n_t) = \frac{1}{n_t t_0} A(t)$$

The triple integral still seems formidable. The integration over aperture can be reserved until last, since the distribution is always perfectly square wave. Thus for the distribution of energy in the plane of the image, one has

$$E_n = \frac{E}{n_0} = \frac{2\pi}{n_0} \iint J_n dn_\sigma \cdot A_n dn_t \cdot I(n, n)$$

One interesting conclusion may already be stated about the distribution in the aperture. Since $I(n)$ is periodic and repeats exactly with each integer, the total energy passed through any annular ring is dependent only on the fractional value of n at the two edges of the aperture. That is, in the multi-ringed pattern of Fabry-Perot fringes, the total energy in each ring is the same, although the diameter and width of the successive rings vary as one moves out from the center. This result is simply due to the fact that the area in the image plane between half maximum points, for example, is the same for all rings.

If the width of the J and A distributions is small compared to the width of $I(n)$, we obtain more rigorously the results stated in an earlier section for the reflectivity limited case. The value of $I(n)$ is then sensibly constant over the range of variation of n_r and n_t , and the integrals are separable.

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$$E_L = \frac{2\pi A B I(R, n)}{n_0}$$

where

$$A = \int A(n_t) dn_t = \int A(t) dt = \text{area of interferometer plates}$$

$$B = \int J_\phi d\phi = \text{integrated line brightness}$$

Now it does not matter how narrow these distributions may be, so long as they are narrow with respect to $I(n)$, the fringe energy will still have the same shape and absolute value. It is further clear that the aperture may be opened up, with increase in the total peak energy and without decrease of contrast, until the peak energy begins to fall off. That is, the proper aperture is the one which just about subtends the half width of the final fringes. The rigorous integral^{*} of $I \delta n$ for this aperture gives: $\frac{\pi}{4} \delta n_R$ and the contrast is reduced from its peak value by $\frac{\pi}{4}$.

* Proof: The integral is

$$\int_{n_a = -1/2 \delta n_R}^{n_a = +1/2 \delta n_R} \frac{1}{1 + m \sin^2(\pi n_a)} dn_a$$

$$\text{where } m = \frac{4R}{(1-R)^2}$$

$$\delta n_R = \frac{1-R}{\pi \sqrt{R}}$$

This integral is found in Pierce's Tables, formula 314. The result is

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$$\frac{1}{\pi \sqrt{R+1}} \left| \tan^{-1} (\sqrt{R+1} \tan \kappa n_R) \right| + \frac{1}{2} \tan_R$$

$$- \frac{1}{2} \tan_R$$

For high values of R , $\frac{\pi}{2} \tan_R = \frac{1-R}{2\sqrt{R}} \ll 1$, and $\tan \theta \approx \theta$. Hence the integral is given fairly closely by

$$\tan_R \frac{2\sqrt{R}}{1+R} \tan^{-1} \frac{1+R}{2\sqrt{R}}$$

From $R = 80\%$ to $R = 100\%$, $\frac{1+R}{2\sqrt{R}}$ is very close to 1 (within .01). The angle whose tangent is one is $\pi/4$ radians. Hence

$$\left. \begin{array}{l} + \frac{1}{2} \tan_R \\ - \frac{1}{2} \tan_R \end{array} \right\} \tan \approx \frac{\pi}{4} \tan_R$$

Thus the absolute available energy and contrast for monochromatic light and flat plates is

$$\left. \begin{array}{l} E = \left\{ \frac{2\pi AB}{n_0} \right\} \frac{\pi}{4} \tan_R = \frac{\pi^2 AB}{2n_0 R} \\ e = 0.31 N_R^2 = \frac{0.31}{\tan_R^2} \end{array} \right\} \tan_R = \tan_R \quad (20)$$

$$(\tan_R = \frac{1}{N_R} = \frac{1-R}{\pi \sqrt{R}} \approx \frac{1-R}{3})$$

Thus one can improve the contrast, but at the expense of energy by increasing the reflectivity toward unity. Eventually of course one must reach the point where the reflectivity width \tan_R is less than either

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the source width or the flatness defect. It is of interest to investigate this case. By a similar argument to the one used above, it is evident that only the wider of the two distributions will be of interest. In our experiments, for example, it is fairly conclusive that the source width is the limiting factor, about 1/12 order, the plates being flat to at least 1/60 order.

The required integral for the total energy is now

$$E = \int_{\text{aperture}} E_n dn_n = \frac{2\pi A}{n_0} \int_{\text{aperture}} \left\{ \int_{n_g} J(n_g) dn_g I(R, n) \right\} dn_n$$

We consider the case when the width Δn_R of $I(R, n)$ is much less than the width Δn_g of $J(n_g)$. We integrate the expression in brackets for two choices of the aperture, first at the peak, then at the minimum midway between fringes. First, at the peak, $I(n)$ varies rapidly while J remains constant. Thus the bracket integral becomes

$$J(n_g) \int_{\substack{\text{whole} \\ \text{fringe}}} I(n) dn$$

But since $I(n)$ has a peak of unity and a half width Δn_R , this last integral has the approximate value Δn_R . The fringe shape is thus J , the source shape. This time the aperture might as well be opened to the half width $\Delta n_n = \Delta n_g$ of the source, and the peak available energy has the approximate value

$$E_{\text{peak}} = \frac{2\pi A}{n_0} J(n_g) \Delta n_g \Delta n_R = \frac{2\pi A^2}{n_0} \Delta n_R$$

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This is the same as obtained in the previous case.

At the minimum of the fringe $I(n)$ changes very slowly, so the bracket integrates to

$$I_{\min} \int J(n_0) dn_0 = I_{\min} B$$

and $I_{\min} = 1/\epsilon_R$, ϵ_R = ideal reflectivity limited contrast and the total energy for aperture $\frac{\partial n_R}{\partial n_0} = \partial n_0$ is

$$E_{\min} = \frac{2\pi AB}{n_0} \frac{\partial n_0}{\epsilon_R}$$

Hence, the effective contrast is

$$\epsilon = \frac{E_{\text{peak}}}{E_{\min}} = \epsilon_R \frac{\partial n_R}{\partial n_0} = 0.4 E_R N_0 \quad (21)$$

Thus the reflectivity contrast is not obtained when $\partial n_R < \partial n_0$.

Conclusions:

- (1) The total energy in each Fabry-Perot ring is the same.
- (2) The total peak energy is in absolute value approximately

$$\frac{2\pi AB}{n_0} \partial n_R = \frac{2}{3} \frac{AB}{n_0} (1 - R) \quad (22)$$

B = source brightness, A = plate area, $\frac{2\pi \partial n_R}{n_0}$ = effective solid angle.

- (3) If the fringe width is purely reflectivity limited, the contrast is

$$\epsilon = 0.4 E_R^2 ; \quad E_R = \frac{\pi \sqrt{N}}{1 - R} \quad (23)$$

- (4) If the fringe is source width limited

$$\epsilon = 0.4 E_R N_0 ; \quad N_0 = \frac{\Delta \phi}{\phi_0} = \frac{1}{25 \phi_0} \quad (24)$$

If flatness limited, replace N_0 by N_t .

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(5) Given a source line width, the optimum resolution, energy and contrast are achieved when the plates are at least the same flatness or better, and the reflectivity and aperture setting satisfy

$$\Delta n_R = \Delta n_A = \Delta n$$

6. TESTING THE FLATNESS OF FABRY-PEROT PLATES

At the start of this project there were a number of quartz plates available which had been prepared by Mr. Wagn Margbol of Arlington, Massachusetts. Mr. Margbol reported that at that time he had no facilities for testing flats to better than $1/50$ fringe of light, but that these plates were at least this flat. Inquiries were made of numerous sources of quartz flats. Hilger in England and Halle in Berlin thought that their plates were flat to at least $1/75$ fringe. Two plates were ordered from Halle. The Hilger plates were considerably more expensive.

It was considered desirable to be able to test flats in our laboratories, in order to test the claims of various producers, in order to determine to what extent the Fabry-Perot resolution would be limited by flatness, and further because it was felt that the local supplier, Mr. Margbol, could make plates as well as anybody if he only had some means to test the progress of his work.

There are a number of methods for testing flats which have been described in the literature. Almost all are based on some form of multiple beam interference fringes produced when the plates are coated with highly reflecting surfaces and mounted with a very slight wedge between them. Two simple optical systems for observing these fringes are sketched

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in Figure 15, one in transmission and one in reflection. In transmission narrow light fringes on a dark background are produced, and in reflection narrow dark fringes on a light background. The transmission method was considered simpler and made use of.

The first tests were made with silver coated plates. The fringes were too wide for accurate determination of flatness. Typical results are indicated in the photograph of Figure 16^a. The width of these fringes seemed to be about 1/10 the fringe separation. There is little observable departure from straightness, but it would be hard to detect better than 1/20 fringe with such wide fringes.

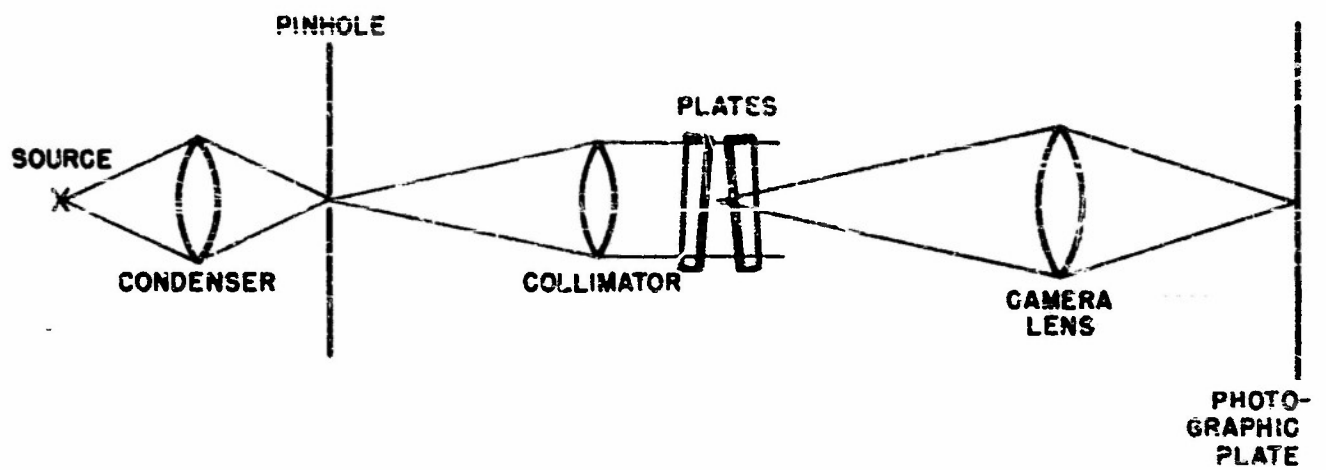
One of the best discussions of the conditions to be met to achieve narrow fringes for plate testing purposes is found in Tolansky's "Multiple Beam Interferometry". It requires multiple beam interference to produce fringes narrow with respect to their separation. The fringes are produced when

$$2\lambda = 2\mu t \cos \theta$$

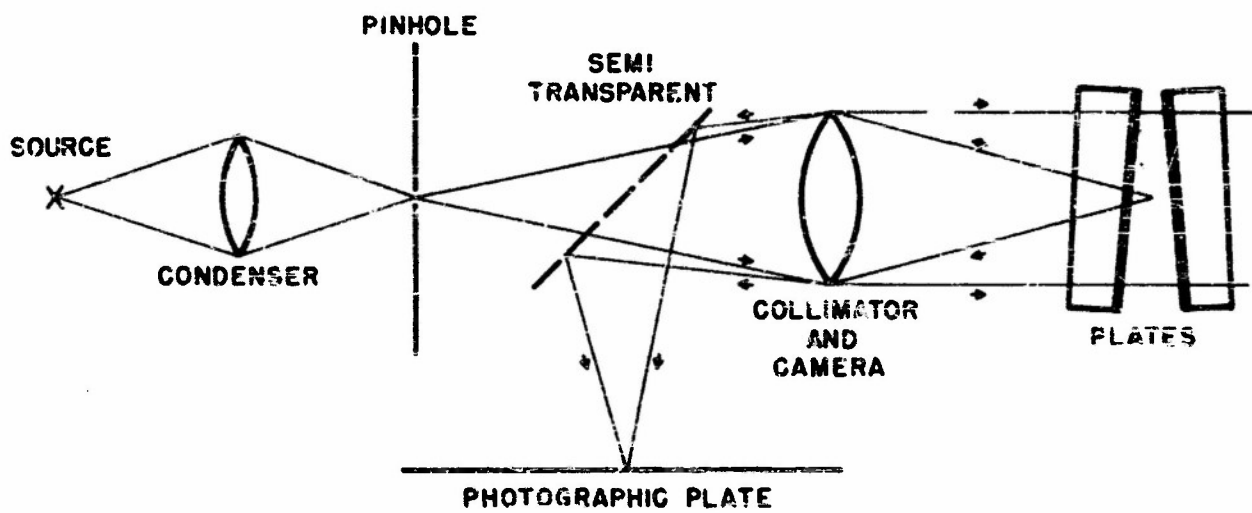
There are three variables (assuming μ constant), t the plate separation, θ the angle of observation and λ the wavelength of the illumination.

If t and λ are held constant we have fringes of equal inclination. These are the Fabry-Perot fringes, with which most of this report is concerned. The transmission varies with θ and the fringes are therefore focussed at infinity.

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* Some of the early work on flatness testing at these laboratories is reported in detail in an earlier technical report, reference 8.
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A. PLATE TESTING IN TRANSMISSION



B. PLATE TESTING IN REFLECTION

FIGURE 15



FIGURE 16.



FIGURE 17.

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If θ and λ are held constant the same fringe-intensity distribution results (equation 5), but the variation with t is observed. These are called fringes of equal thickness and the fringes are focussed approximately in the plane of the plates. These are the fringes then that are useful in observing the flatness of plates.

The third type, with t and θ constant, λ variable, Tolansky calls fringes of equal chromatic order.

Actually, in the pressure tuning method, the fourth variable μ is used.

There are four conditions which must be satisfied in order to obtain the sharpest fringes.

(1) The fringes cannot be narrower than the reflectivity limit discussed above

$$\delta n_p \approx \frac{1-R}{3}$$

(2) The absolute separation between the plates must be kept very small. This is because of the wedge introduced to produce the series of fringes across the face of the plate. As a result the phase change for each successive beam in the multiple interference is not the same, as in the perfectly parallel case. By detailed consideration of this effect Tolansky shows that the absolute plate separation must be less than

$$t < \frac{1.5(n_R)^3}{\lambda X^2}$$

where X is the number of fringes per centimeter across the wedge. When there is no wedge (Fabry-Perot) then t can be large (i.e., X is small).

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(3) Above it was stated that θ is to be constant, i.e., parallel light. More detailed consideration shows that the collimation must be at least

$$\Delta\theta < \sqrt{\frac{\lambda n_B}{9t}}$$

(4) The source width must of course be less than the reflectivity width.

$$\Delta x < \frac{\lambda^2}{9t} \tan \theta$$

In the first attempts the principal difficulty was with the poor reflectivity of the silver. This was remedied by coating the zinc plates with nine multilayer films, having a reflectivity of about 97%. This gives a theoretical fringe width of .01 fringes. The other three conditions were met, and the fringes shown in Figure 17 were obtained. These are by the optical arrangement of A, Figure 15.

These fringes were quite sharp. It was estimated that the width was at least 1/80 fringe. On the original plates fine microscopic wiggles can be detected which are presumed to be the microscopic roughness of the plate. The central portion of the plate seems to be flat to around 1/80 fringe. There is a pronounced rounding of the fringes toward the edge of the reflecting layers, which were about 1.4 inches diameter on the 2 inch plates. It is not known whether this rounding off is due to lack of plate flatness or whether it may be caused by non-uniformity of the evaporated multilayers. Further study of the uniformity of laying down nine dielectric layers would be required. It was evident however

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that for the central portion of the plates, the flatness would be less of a limitation on the hydrogen-deuterium analysis than the actual width of the hydrogen lines in the source.

7. IMPROVING ELECTRONIC SIGNAL-TO-NOISE

The work on the testing of flatness and reflectivity in the Fairy-Parot plates was now convincing that the resolution was limited only by the actual source width. What was being observed was the Doppler broadened fine structure of the H- α line, the source broadening being so large that the seven actual fine structure components¹ were observed merely as two broad lines just barely resolved, that is, highly overlapped.

At this time recorder traces as indicated in Figure 18 were being obtained. The contrast, that is, the ratio of the peak H intensity to the minimum H-fringe intensity at the D position, was about 240. (Dark current and room stray light were of course subtracted out.) The sample for Figure 18 was prepared to approximately a deuterium concentration of 0.5%, and the ratio of the D-light to the H-light was approximately this value.

It was apparent that further significant gains in pushing the concentration sensitivity to lower deuterium concentrations would be obtained only by improving the electronic technique of reading the D peak in the presence of the H-background. Now in such situations, it is obvious that the background itself, if it were constant, is no obstacle to infinite sensitivity for it can be merely subtracted out. What will ultimately limit the sensitivity is the fluctuations in the background. Theoretically, if one could eliminate all other sources of noise, the

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1 The fine structure and Doppler broadening of H α and H β are discussed in the preceding Technical Report under this contract, dated July 1, 1953.
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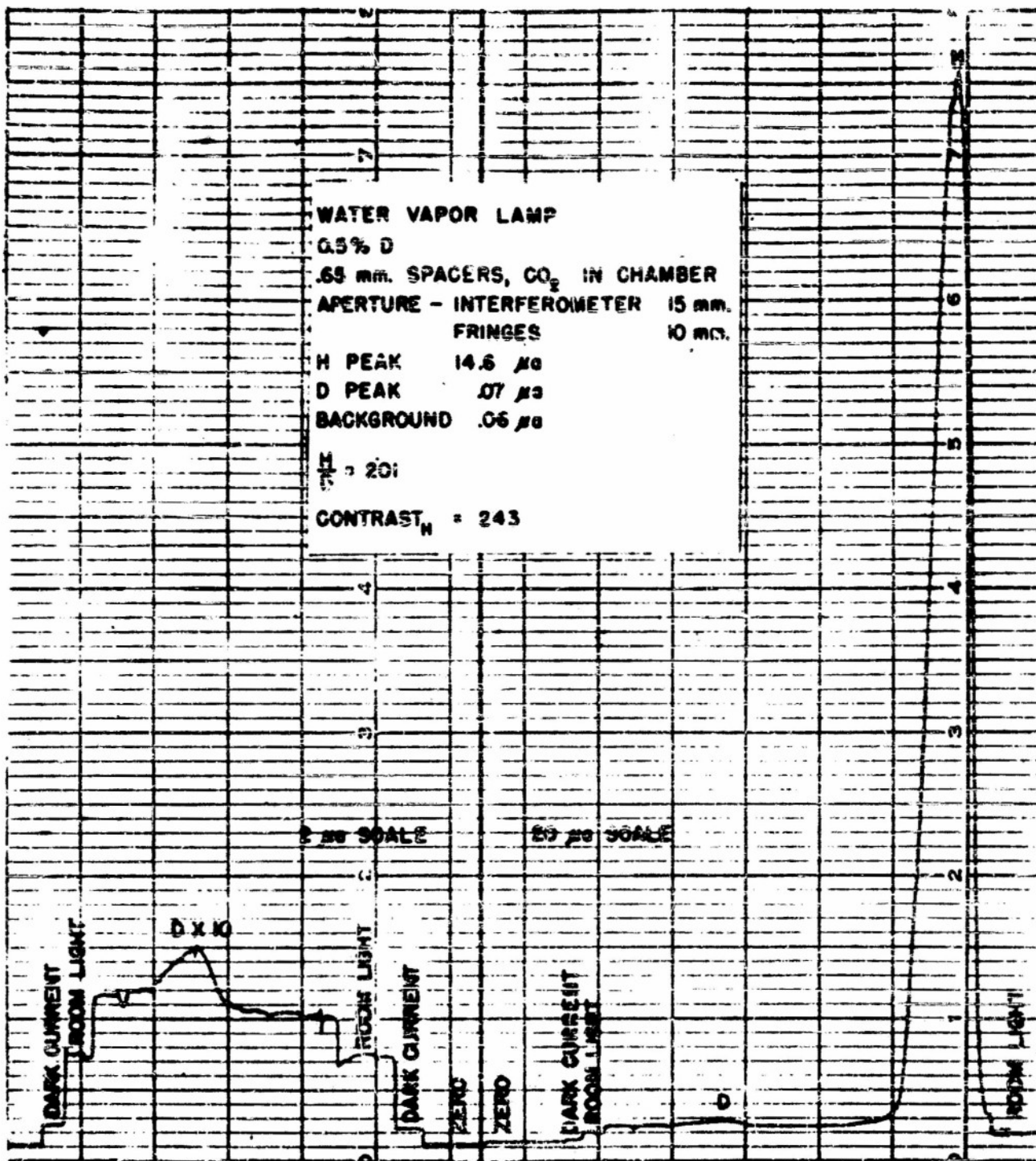


FIG. 18

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ultimate limit would be set by the shot effect fluctuations in the electrons emitted from the cathode of the photomultiplier. The magnitude of this effect is well known^{*}.

The rms fluctuation noise in the cathode current is given by

$$(\Delta I_k)_{\text{rms}} = \left(\frac{I_k e}{\tau} \right)^{1/2}$$

where I_k is the average cathode current, e the charge of the electron, and τ the time constant of the overall detector-indicator. The current I_k can of course be expressed as the product of the available light flux F by the absolute sensitivity S of the photo-cathode, and besides these two factors, only the time constant influences the noise level. Thus one can increase the signal-to-noise ratio only by increasing the light flux, the photo-cathode sensitivity or the time constant.

$$\frac{S}{N} = \frac{I_k}{\Delta I_k} = \left(\frac{I_k \tau}{e} \right)^{1/2} = \left(\frac{F S \tau}{e} \right)^{1/2}$$

In the photomultiplier the gain of the dynode stages is simple amplification of the noise and signal by the same factor G , so that the signal-to-noise is not changed in the anode current. However since the cathode current is not readily measured, it is perhaps convenient to record the noise equations relating to the anode current I_A .

$$(\Delta I_A)_{\text{rms}} = G(\Delta I_k)_{\text{rms}} = \left(\frac{G I_A e}{\tau} \right)^{1/2}$$

$$\frac{S}{N} = \frac{I_A}{\Delta I_A} = \left(\frac{I_A \tau}{e G} \right)^{1/2}$$

* See, for example, R. W. Engstrom, J. Opt. Soc. Am. 37, 420 (1947) or R. O'B. Carpenter, J. Opt. Soc. Am. 40, 225 (1950).

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This has the same numerical value as the expression obtained above, but it is usually laborious to measure the photomultiplier cathode current and absolute sensitivity. With the last expression one must estimate the gain G . A rough value can be obtained from the RCA handbook, but too much reliance should not be placed on it because of the tremendous variation in gain from sample to sample.

The above expressions for S/N assume that the entire current consists of the signal. When there are extraneous currents added to the signal, such as photomultiplier dark current or room stray light, the above expressions for S/N are to be multiplied by the ratio of the signal current to the total current.

Therefore the photomultiplier and recorder system were critically investigated in order to determine the limiting noise factors and the ultimate concentration sensitivity which could be obtained in hydrogen-deuterium determinations. The hope was to be able to reach the detection of the deuterium existing in ordinary water, namely .015 percent.

A first difficulty appeared because the maximum scale sensitivity of the Leeds and Northrup Speedomax being used was 2 microamperes full scale. This strip chart recorder is "dead" for deflections less than about 0.2 percent of full scale. With this scale and 1000 volts on the photomultiplier it was just barely possible to observe fluctuations in the trace. Therefore it was desirable to increase the sensitivity of the recording system. This could be done in two ways. The photomultiplier voltage, and hence its gain G , was increased. Soon the safe limit of 1250 volts was reached. The input circuit of the Speedomax was investigated to see whether

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its sensitivity could be increased. It was found that a simple change would give us a factor of 2, or a 1 microampere full scale setting. It appeared that any further gains would be time-consuming in that the entire coupling and damping input network of the recorder would have to be completely redesigned.

Although we would have preferred to be able to go further with the increase of gain, considerations of time and expense precluded this, in view of the fact that with these simple changes fairly considerable noise fluctuations were now detected. These are shown in Figure 19.

The first trace shows a current of approximately 0.75 microamperes on the 1 microampere scale. This current was derived from a dry battery and resistance and shows that there is no noise introduced by the recorder. The recorder also showed no dead spot or overshoot within 0.2% full scale. The next trace is a light signal from a tungsten lamp operated from a storage battery, which is known to give a highly stable and noise-free light flux. The gradual drift in current may be fatigue of the photocell. At any rate, the noise fluctuations are quite large. The succeeding trace shows the reduction of this noise when the time constant of the recorder is increased by simply connecting two 8 μ f oil capacitors in parallel across the input leads. The effective input resistance of the recorder is about 70,000 ohms so that the time constant can be estimated from the RC value. With no external capacitance the time constant is about 1 second, being limited both by some internal capacity and by the inertia of the motor-servo system.



FIG. 19A

0.75 μ s FROM DRY BATTERY.
DEAD SPOT TESTS

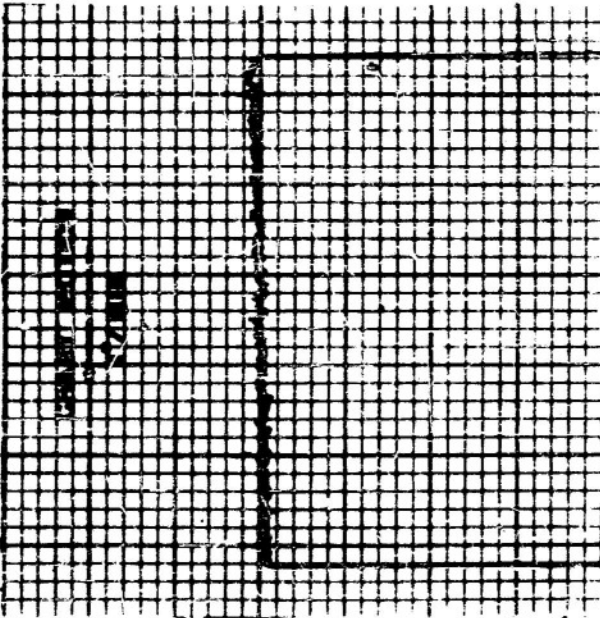


FIG. 19B

0.75 μ s FROM PHOTOMULTIPLIER.
1250 V ON PHOTOMULTIPLIER AND
BATTERY OPERATED TUNGSTEN LAMP

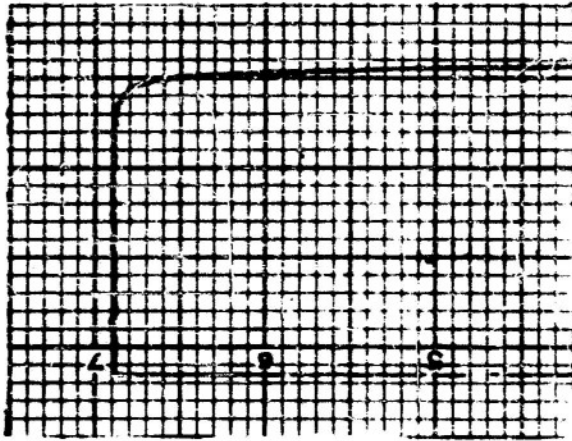


FIG. 19C

16 μ s ADDED ACROSS IN-
PUT FOR 19B

TESTS ON 1 μ s SCALE OF RECORDER

FIG. 19

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8. FINAL EVALUATION OF PERFORMANCE

Three other sources of noise were noted at various times. Variations in photomultiplier gain due to line voltage fluctuations were practically eliminated by use of a Sorenson voltage stabilizer.

Stray room light became significant when attempting to measure low concentrations. Most of the light reaching the photomultiplier seemed to be from reflection off the vacuum chamber holding the interferometer. The unwanted light reaching the photomultiplier was reduced to a negligible amount by putting the photomultiplier in a long box with a variable iris just in front of the interferometer focussing lens. This iris also acted as the limiting aperture for the Fabry-Perot interferometer.

The microwave exciter used with the lamp was also operated from a voltage regulator but the magnetron output still had an occasional dip which results in a sharp intermittent pip on the recorder trace. This does not interfere seriously with the measurements but is certainly not desirable.

After the noise reduction work a sample of approximately 0.05% D was prepared.

Two limitations of the quickly assembled equipment used for these preliminary studies became apparent. First the interferometer spacers are 0.65 mm thick. This gives a difference in order between I_x and D_x of 0.54 order instead of the difference of 0.50 order necessary to put D maximum at the I minimum. For an 0.05% concentration of D and a contrast of 200 the ratio of D_{max} to I_{min} is

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$$\frac{D_{\max}}{H_{\min}} = \frac{\frac{H_{\max}}{2000}}{\frac{H_{\max}}{200}} = 0.1$$

Since the H fringe increases by a few percent over its minimum at .54 the D maximum is seen as just a bulge on the side of a sloping line. Since the control of pressure change is rather crude the recorded traces are not linear with pressure (or order). The rate of change of pressure can cause an irregularity in slope which may be discouragingly similar to the D maximum as in Figure 20.

The adjustment to parallelize of the interferometer plates is closely related to this difficulty. The parallelism of the plates is the only factor affecting the contrast which is likely to change in the short time of a few days or weeks. Besides the obvious effect of making the D_{\max}/H_{\min} ratio smaller it also changes the shape of the fringes apparently in a way that is not helpful. The trace in Figure 20 was for a contrast of about 130 and a D concentration of 0.05%. The trace in Figure 21 is for the same conditions except that the interferometer had been adjusted so that the contrast was 210. Here the D_{\max} can be seen quite easily and D_{\max}/H_{\max} is about 0.05%. However the slope of the H background makes the measurement of the D peak rather difficult. It is definitely of great importance to obtain ball spacers of the exact required thickness. A manufacturer will probably have to be asked to make a special run.

There was not enough time to make a series of runs with natural water but one test seems to indicate that the 0.015% D concentration is detectable with the present equipment although it cannot be measured with much accuracy.

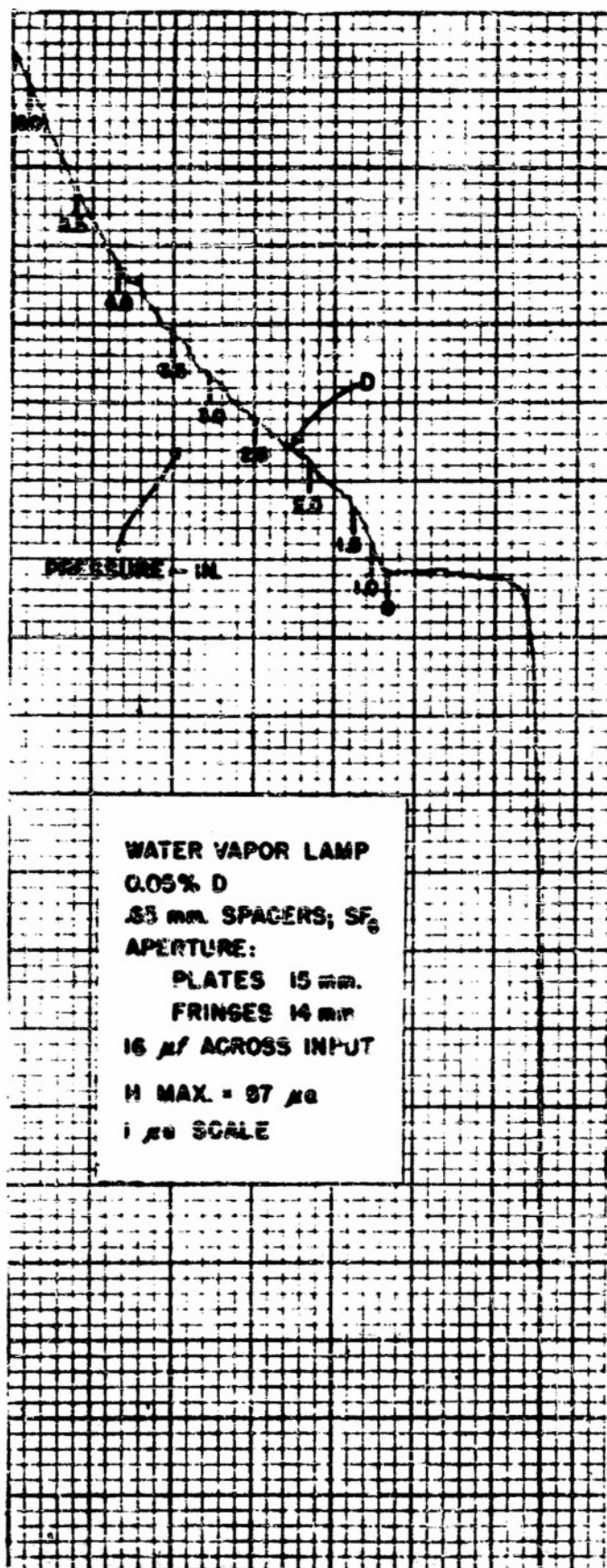


FIG. 20

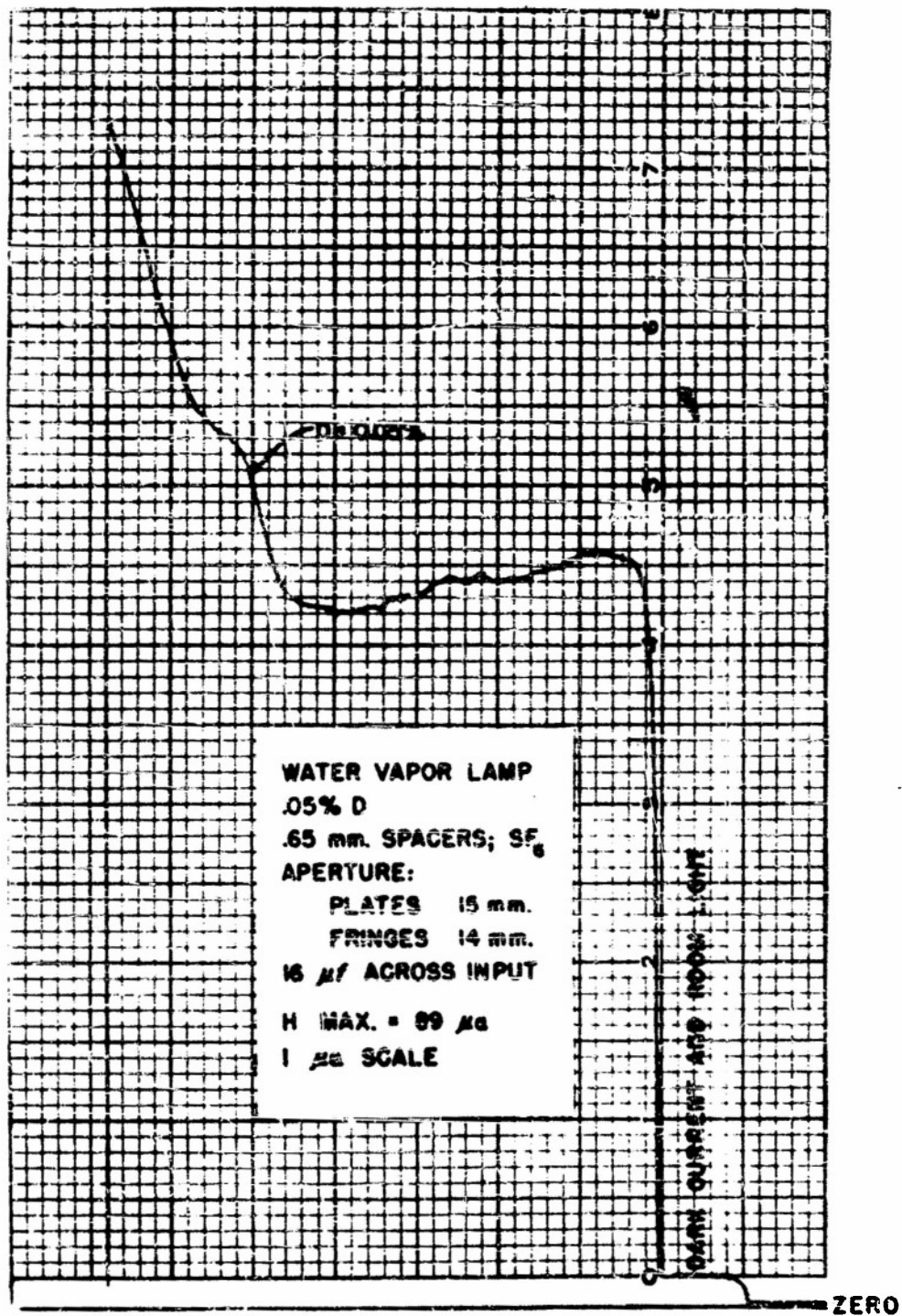


FIG. 21

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9. COMPARISON OF THE LUMINOSITIES OF FABRY-PEROT, GRATING, PRISM AND POLARIZATION INTERFEROMETER

In evaluating a variety of dispersing instruments for use in spectrochemical analysis, one of the points of interest will be the relative light gathering power of the various instruments, sometimes called the "luminosity". This is particularly true when one wishes to measure quite low concentrations of an element in a source which is inherently not particularly bright, in other words, in an energy limited situation. The luminosity may then determine whether the electronic noise is limited by the photon shot noise, or by noises originating in the detector-amplifier combination.

The general formula for the energy gathering power of any optical system is

$$E = BA\omega T$$

where B is the source brightness

A is the area of the optical beam

ω is the solid angular divergence of the beam

T is the relative transmission of the system as limited by absorption or reflection.

This formula will be applied in turn to the Fabry-Perot interferometer, a grating spectrograph, a prism spectrograph, and the polarization interferometer. Since B refers to the source, and may be assumed to be the same for competing dispersing elements, the figure of merit of the dispersing elements will be the luminosity, defined as

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$$L = A n T$$

This quantity will have the dimensions of area.

The luminosity of any instrument will depend on the resolution which has to be achieved. It will be assumed that the problem requires a resolution $R = \lambda/\delta\lambda$, and that all the source energy is in a band smaller than $\delta\lambda$.

1. The Fabry-Perot Interferometer

The work of calculating the aperture and luminosity of the instrument has already been performed in the preceding section. The result for the total energy passed was

$$\frac{\pi^2}{2} \frac{A^2 B}{n_c^2}$$

However in an earlier section of this report, $n_c R$ was found to be the resolution R of the system. Hence the luminosity of the Fabry-Perot is given by

$$L \approx \frac{A^2}{R} \quad (25)$$

Note: the factor $T_M = 1 - \frac{A}{(1-R)}$ has been left out on the assumption that non-absorbing multilayer reflectors are used. For silver or other absorbing reflectors this factor should be included.

2. The Grating Spectrograph

For the grating spectrograph the solid angle of the grating as seen by the exit slit is

$$\frac{A}{r^2} \cos r$$

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where A_g is the area of the grating rulings, r the angle of diffraction and f the focal length.

The transmission factor of the grating varies considerably from grating to grating, and with wavelength, order, blaze, etc, but for a properly chosen well-blazed grating might easily be about 30% at H alpha.

The area of the slit is of course simply its width times its length. The width is limited by the resolution desired and the dispersion. The dispersion is given by

$$n_0 \lambda = a(\sin i + \sin r)$$

$$\frac{d\lambda}{dx} = \frac{1}{f} \frac{da}{dr} = \frac{a}{fn_0} \cos r$$

where r is the angle of diffraction, n_0 is the order number, a is the grating line space constant. To achieve resolution $R = \lambda/\Delta\lambda$, we must use a slit width

$$\Delta x = \Delta\lambda \frac{dx}{d\lambda} = \frac{\lambda}{R} \frac{dx}{d\lambda} = \frac{\lambda^2 n_0}{R a \cos r}$$

$n_0 \lambda/a$ can be eliminated by means of the grating formula above

$$\Delta x = \frac{f}{R} \frac{(\sin i + \sin r)}{\cos r}$$

The length l of the slit is commonly of the order of 1 inch. It is limited usually by the fact that above or below the plane of the Rowland circle, the astigmatic image lines become not perpendicular to the Rowland circle nor parallel to the slit. For this reason Fastie¹³ devised a type of grating mount originally proposed by Ebert with circular

13. William G. Fastie, J. Opt. Soc. Am. 42, 641, 647 (1952).

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rather than straight slits, with entrance slit and exit slit symmetrical with respect to the grating, in which the slit length could be increased to 4 inches and more without astigmatism troubles. Hence the final result for the luminosity is

$$L = A_{\text{eff}} = \frac{T}{R} \frac{A}{f} \frac{1}{2} (\sin i + \sin r)$$

This result is less than the luminosity of the Fabry-Perot by approximately 1/50. Ordinarily one would expect about $f = 40''$ (1 meter), $i = 1''$, or the grating is about 1/200 as fast. As pointed out above, astigmatism usually restricts the slit length.

3. The Prism Spectrograph

In the case of the prism the derivation will be omitted. But in the case of minimum deviation, which is a condition for maximum luminosity, the result can be written:

$$L = \frac{2A}{R} \frac{1}{f} \frac{\lambda}{n} \frac{dn}{d\lambda}$$

The luminosity is again limited by the slit length factor, and also by the final factor depending on the dispersion of index. The area A_{pr} in the above result is the area of one of the faces through which the light passes. Typical values for the dispersion factor $\frac{\lambda}{n} \frac{dn}{d\lambda}$ are between 0.1 and 0.01. No absorption losses are included in the above formula.

4. Polarization Interferometer

In the last technical report submitted under this project (dated July 1, 1953) an interesting instrument for separating E and D light was discussed, based on the dispersion of birefringence. This was

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called the polarization interferometer. The light was not actually dispersed in ordinary space, but the H and D light was polarized at right angles to each other. In this section the luminosity of this system is compared with those computed above.

This comparison is included in this report for completeness, and also because of the interesting properties of the polarization interferometer. The interested reader may have to refer to the detailed discussion of the last technical report for the definition of all the terminology.

If polaroids are used for the three polarizers their transmission will be about $.50(.70)^3 = 17\%$. 50% is lost in the initial polarization of the light. In order to get a high percentage polarization, fairly dense polaroids must be used, so that the transmission for the parallel component will probably not exceed 70%. Hence

$$T_p = 17\%$$

Other sources of loss in transmission will be reflection losses, glass and crystal absorption. These will be small if the system is immersed in oil, which will probably be necessary for temperature control purposes anyway.

The angular field of these x-cut crystal plates has been described by Evans¹⁴. For the simple plates without the field widening technique the phase retardation as a function of the angle of transmission through the crystal is given by

14. John W. Evans, J. Opt. Soc. Am. 39, 229 (1949).

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$$\gamma = \gamma_0 \left\{ 1 + \frac{\theta^2}{4n_0^2} \left(\frac{\cos^2 \theta}{n_o} - \frac{\sin^2 \theta}{n_e} \right) \right\}$$

$$= \gamma_0 \left\{ 1 + \frac{\theta^2}{4n_0^2} \right\} \quad (\text{at } \theta = 0, 90, 180, 270)$$

where θ , ϕ are the polar coordinates of the angle of incidence, n_o , n_e the ordinary and extraordinary refractive indices of the uniaxial crystal. For the Lyot-Evans type field widened system (two plates of half thickness separated by a half wave plate)

$$\gamma = \gamma_0 \left\{ 1 + \frac{\theta^2}{4n_0^2} \left(\frac{1}{n_o} - \frac{1}{n_e} \right) \right\}$$

Hence the minimum improvement by the field widening (i.e., ratio of angles of incidence having same $\Delta\gamma$) is

$$\left(\frac{n_o}{n_o - n_e} \right)^{1/2} \approx \begin{array}{l} 8 \text{ for AIR} \\ 41 \text{ for calcite} \\ 18.2 \text{ for quartz} \end{array}$$

The above formulas give the deviation in retardation for the ray deviating from the normal by θ degrees. If we consider a circular cone of rays having a maximum angle of incidence of θ' , the average deviation in retardation is computed as follows:

$$(\Delta\gamma)_{av} = \left(k/\pi\theta'^2 \right) \int_0^{\theta'} r^2 2\pi r dr = \frac{1}{2} k\theta'^2$$

$$k = \frac{\gamma_0}{4n_0^2} \left(\frac{1}{n_o} - \frac{1}{n_e} \right) \quad (\text{corrected plates})$$

In other words, the average $\Delta\gamma$ is one-half the maximum $\Delta\gamma$ for a circular

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cone of rays. The solid angle ω_p of this cone is

$$\omega_p = \pi \theta^2$$

Since over the cone of divergent rays, δ varies from its mean value to $\delta_0 \pm (\Delta\delta)_{av}$. The solid angle permitted, and hence the light gathering power depends on the permitted variation in $\Delta\delta = \Delta\theta$.

$$\omega_p = \frac{4\pi n_o^2 n_e}{n_e - n_o} \frac{(\Delta\delta)_{max}}{\delta_0}$$

$$\frac{4\pi n_o^2 n_e}{n_e - n_o} = \begin{array}{l} 967 \text{ for AIF} \\ 300 \text{ for calcite} \\ 5,150 \text{ for quartz} \end{array}$$

Since the resolution R will be $\delta_0/\Delta\delta$, the general result for the luminosity will be

$$L = \frac{T_p A}{R^2} \frac{4\pi n_o^2 n_e}{n_e - n_o}$$

With $T_p = 0.17$ the ratio of the luminosities of polarization and Fabry-Perot interferometers is approximately

$$\frac{0.17}{5} \left\{ \frac{4\pi n_o^2 n_e}{n_e - n_o} \right\}$$

33 for AIF

10 for calcite

173 for quartz

The luminosity of the polarization interferometer is thus greater than that of the Fabry-Perot, which is in turn more luminous than the grating

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or prism. It was pointed out in the detailed discussion of the last technical report that the disadvantage of the polarization method was that for high accuracy it was restricted to two perfectly monochromatic lines. The higher luminosity may serve to rescue this instrument for practical use where luminosity is a heavy requirement, where the lines are quite narrow, and where the highest accuracy and contrast are not required.

10. SOURCE AND SAMPLING METHODS FOR DEUTERIUM ANALYSIS

In order to use the spectrochemical analysis method on hydrogen-deuterium mixtures, the sample may be in the form of either hydrogen gas or water. Either the gas or the water vapor may be excited in an electrodeless discharge. The electrodeless discharge in a glass tube is preferred because of the possibility of contamination of metal electrodes. Considerable time might otherwise be lost in cleaning out the discharge tube between samples.

Since only a few mm of vapor or gas in a small discharge vessel is required, only a minute amount of sample is required. This is one of the advantages of spectrochemical methods over other methods of analysis.

Apparatus for handling either type of sample have been constructed in our laboratories. The final choice probably depends most strongly on the actual form in which the sample for analysis is directly available. For the medical tracer applications the sample is usually liquid water. One or two drops suffice for the determination. The apparatus for the water vapor discharge is somewhat simpler than that for the gaseous discharge, but this is only if provision for dilution of samples for calibration, or otherwise, is required.

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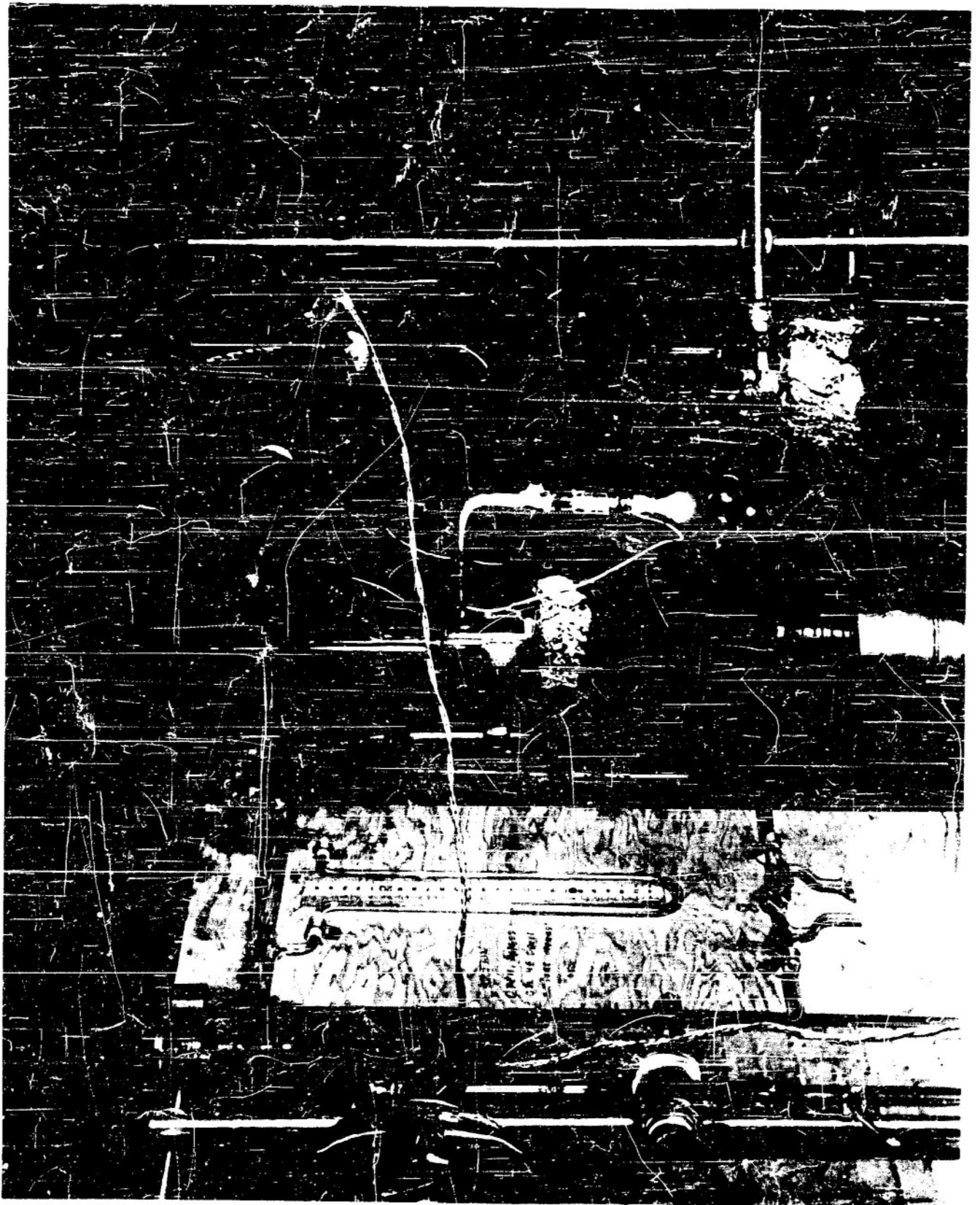
The apparatus for mixing and discharging hydrogen and deuterium gases has been described in detail by Broda and Moyer of the National Bureau of Standards (J. Opt. Soc. Am. 42, 37 (1952)), who made an extensive investigation of the intensity ratio with pressure and with exciter intensity. Our apparatus was somewhat similar to their design, except that only one palladium tube was available. The photograph of our apparatus is shown in Figure 22.

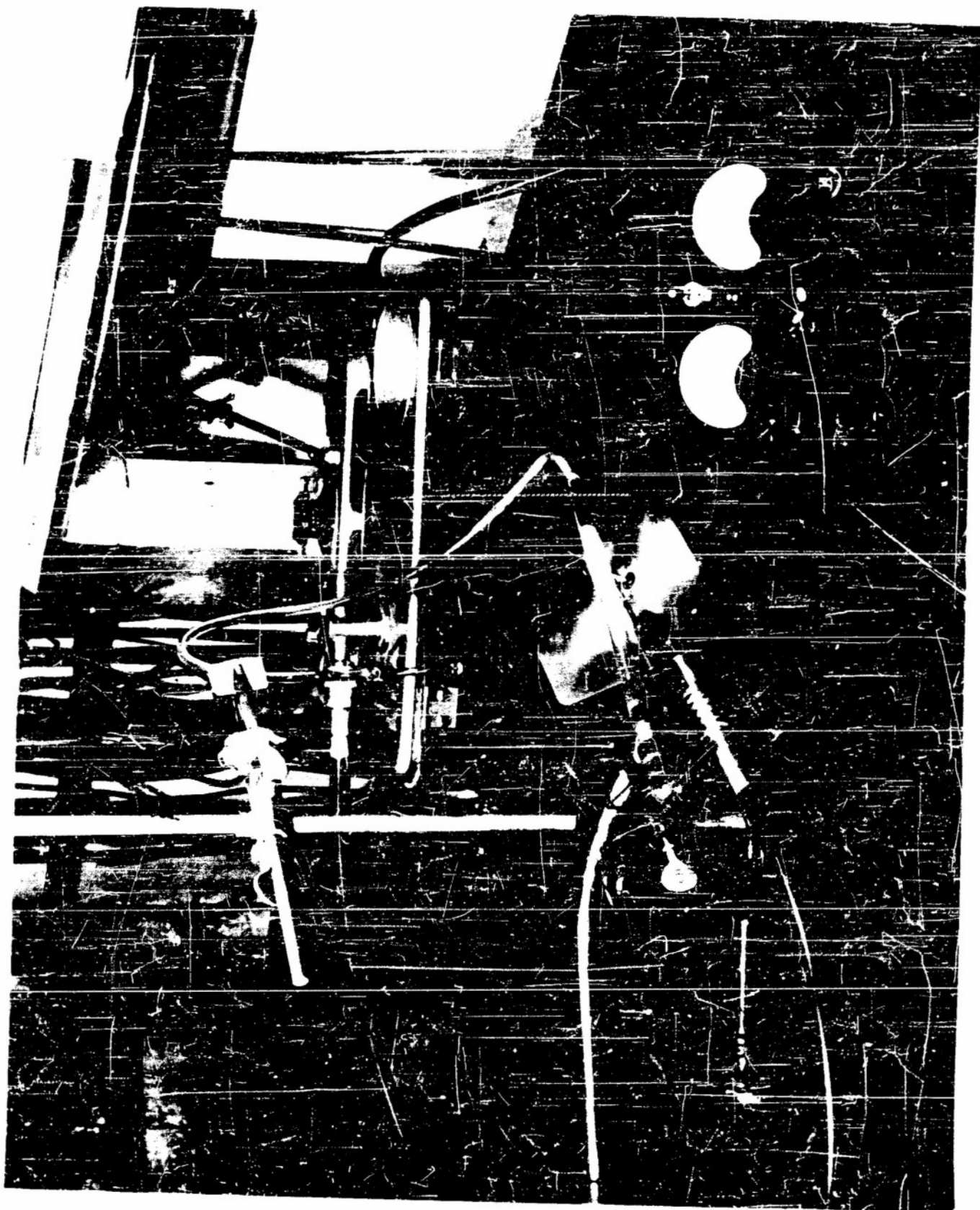
The convenience of the water vapor method induced us to do most of the testing of the interferometer with this source. The apparatus is shown in Figure 23. A 125 watt microwave magnetron generator at 10 cm wave length was used for the excitation of the discharge.

11. IMPROVEMENT OF CONTRAST

If one attempts to measure deuterium down to the low concentration available in ordinary hydrogen, a rather extreme contrast of 8000 to one is required. By careful optimization of the ordinary techniques of Fabry-Perot interferometry it has been just barely possible to achieve this contrast. Since this problem may occur in even greater severity in similar isotope measuring problems, or other applications, it is useful to inquire whether there are available any techniques which will push the contrast to even higher limits. Only two such methods have so far been unearthed, and these were suggested to the author by Professor Jacquinot.

1. Self-Absorption. If light, after being emitted, passes through a long path of the same gas, it may be absorbed. In the case of the strong and weak lines, where high contrast is needed, the self-absorption will be much greater for the strong line. Therefore after traversing the path,





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the two lines will be more nearly equal in intensity, and there will be an apparent decrease in the ratio of the light intensities. The situation in H-alpha is rendered more difficult because the final state is not the ground state, therefore the line is not considered to be one susceptible to a high degree of self absorption. That is, before the absorbing path can absorb, there must be present a high concentration of hydrogen atoms in the first excited state. This means that a vessel of hydrogen gas alone will not serve as the absorber, but that the absorbing gas will have to be electrically excited as well as the source. Jacquinot has constructed long discharge tubes with multiple reflections in which the ratio of D-alpha light to H-alpha was considerably increased. The amount of self-absorption, of course, will have to be accurately controlled if one is to be able to arrive at a measure of the concentration ratio from the intensity ratio. One of the beauties of the spectrochemical method of H-D analysis was that the intensity ratio was almost exactly the same as the concentration ratio, so that extensive calibration was not required.

2. Interferometers in Series. If the required contrast cannot be obtained with a single interferometer, the use of two interferometers in series should give an overall contrast equal to the product of those obtained with the single instruments. The difficulty here is in maintaining the tuning of the two instruments so that they are simultaneously on the peaks of the fringes. The etalon spacing of course has to be maintained to a fraction of a wavelength of light. The air pressure tuning method was very useful in the single interferometer case. Probably in order to use two interferometers, they should be installed in separate vacuum tanks with independent control.

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Application of this double monochromator principle is not of course limited to two identical instruments in series. A Fabry-Perot could be used in series with a prism, grating or other dispersing instrument of sufficient resolution. One of the best suggestions along this line appears to be a fore-dispersing element consisting of a solid mica-spaced Fabry-Perot plate, permanently tuned to the D-wavelength. After passing through this device, the intensity ratio presented to the Fabry-Perot would be much reduced, and such high contrast would not be required. Quartz is a possibility for this solid spacer fore-disperser. About 0.015 inches thickness is required (air spacer thickness divided by the index) to separate H and D by exactly $1/2$ order. The advantage of quartz is its insensitivity to temperature, humidity or chemical attack. However, lithium or calcium fluoride is probably more desirable because of lower index, hence greater thickness, and because they are available in large synthetic pure crystals at low cost, and are isotropic media. The extremely thin plates required raise problems in standard finishing and polishing methods. Mica would be easier to prepare, but might suffer from considerable absorption in such thicknesses.

Another possibility for the fore-dispersing element is one section of the polarization interference filter described in the preceding report. This could be of Al_2O_3 , calcite or quartz.

If an adequate fore-dispersing element of the type suggested above can be manufactured simply and cheaply, an immediate suggestion is to dispense with the air spaced pressure tuned interferometer and use two such devices in series. The reference beam for the measurement could be

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taken from the undispersed source, with a single narrow band multilayer interference filter used to select the H and D light. After calibration with known samples, a direct reading method would be provided without necessity for scanning, nor for adjustment of parallelism of the plates.

The development of an adequate technique for manufacturing solid spacer interferometers to the required tolerances in mica, quartz or the fluorides suggests an important proposal for future experimental work in this field of high resolution interference spectroscopy.

12. PROPOSALS FOR FUTURE WORK

The report on the hydrogen-deuterium analysis problem by means of the Fabry-Perot interferometer has demonstrated that this instrument has the possibility of the highest luminosity, resolution and contrast for the spectral analytical determination of isotopes. The hydrogen - deuterium isotope shift is of course one of the largest. It is proposed that future work be aimed directly at separating other isotopes of much narrower isotope shifts, notably uranium, because of its importance.

With the heavier elements, although the isotope shift is smaller, the Doppler broadening would be less. Therefore one of the major limiting factors will be the flatness of the interferometer plates. Considerably more work needs to be done both on the techniques of preparing flat quartz plates and on the techniques for testing them. The testing is necessary to the preparation, since the precise optical worker can make fine corrections if he is given very precise information as to where the defects lie. Baird Associates is fortunate in being able to obtain the services of Mr. Wagn Hargbol for this work. The problems have already been discussed,

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and Mr. Margbol is anxious to acquire the best apparatus and techniques for achieving the ultimate in such plates.

Further work needs to be done on the improvement of the techniques of laying down multilayers. This involves purifying the dielectrics evaporated, extensive gadgetry for rotating plates in the evaporator to insure uniformity, solution of the exact theoretical equations for the variation of transmission with thickness of deposited layer so that the optimum monitoring system may be devised, etc.

Another major problem requiring extensive research work is the problem of sources for the heavier elements, such as uranium. The vapor pressure is low, and it is difficult to excite them, even in arcs or sparks. In addition, such arcs and sparks are notoriously sources of extreme fluctuations in light intensity compared to gaseous discharges. Hence we may expect the problem of noise to be intensified with these elements.

An additional complication may be introduced because of the complexity of uranium spectra. For H-D, a 50 angstrom wide multilayer interference filter is quite adequate as an isolating filter. For uranium, it may be necessary to do research on the development of more narrow isolating filters.

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APPENDIX I

THE DETERMINATION OF HEAVY WATER BY INFRARED ABSORPTION TECHNIQUES

By W. A. Patterson

SUMMARY

Using 0.5 mm Calcium Fluoride cells, the differential method of infrared analysis, a reversed cell technique and wide fixed slits, Deuterium Oxide as an additive in water can be readily detected in percentages as low as 0.01% with a probable precision of $\pm 0.001\%$. Under present conditions an ultimate sensitivity of 0.001% Deuterium Oxide seems probable. The determination of ultimate sensitivity has been restricted by bad scattered light conditions when spectrophotometric slits are widened. If this scattered light problem could be solved, then an ultimate sensitivity of 0.001% Deuterium Oxide does not seem out of the way.

Introduction

Sensitive methods for the determination of heavy water (Deuterium Oxide) in the presence of ordinary water have been based either on the mass spectrometer or some light dispersing instrument such as an emission spectrograph or monochromator using electrodeless gas discharges. Thomas¹⁵ describes a mass spectrometric method for D_2O in H_2O which appears to be accurate to $\pm 0.2\%$ in the range 0.2% to 1%, and $\pm 0.3\%$ in the range 1% to 10%. This method is based on the determination of the relative concentrations of mass 19 from DOH and mass 18 from H_2O . Thomas indicates that the $\pm 0.2\%$ limit of accuracy might be due to adsorption of heavy water on the vacuum system walls. This had been realized by Orchin, Wender and Friedel¹⁶

15. B. W. Thomas, Anal. Chem. 22, 1476 (1950).

16. O. Orchin, I. Wender and R. A. Friedel, Anal. Chem. 21, 1072 (1949).

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who, in order to get increased accuracy, converted solutions of D_2O and H_2O to Methyl Deuteride (CH_3D) and Methane (CH_4) before using the mass spectrometer. This method gave values for heavy water to within 0.1% in the range 1.4 to 15%, but neglecting the errors inherent in the "conversion", the reproducibility of the mass spectrometer values on the Methyl Deuteride was at least a factor of 2 better than this.

The Orchin et al method is interesting in that it was applied to mixtures of hydrocarbons of relatively high molecular weight, some compounds of which had Deuterium atoms substituted for Hydrogen atoms. These hydrocarbons were accurately converted to water and Deuterium Oxide by a method developed by Keston, Rittenberg and Schoenheimer¹⁷. This technique has important applications and means that a good method for determining D_2O in H_2O could have wide spread applications.

The papers of Broida^{18, 19} appear to be the most authoritative using emission techniques. In the first paper¹⁸, gaseous mixtures of Hydrogen and Deuterium from 85% to 100% Deuterium were analysed to within 0.1% of the major component. In the second paper¹⁹, with the same instrumentation, and applying isotope dilution techniques the total water content of materials such as "hydrates" was determined to within 3% of the amount present. This paper illustrates at least one other type of application for methods determining D_2O in H_2O . According to the authors, the technique is perfectly general, and applications were planned for the study of water binding and transport in biological systems.

17. A.S. Keston, D. Rittenberg, and R.J. Schoenheimer, J. Bio. Chem. 122, 227 (1957).

18. E. P. Broida and J. W. Moyer, J. Opt. Soc. Am. 42, 37 (1952).

19. E. J. Morawitz and E. P. Broida, Anal. Chem. 24, 1637 (1952)

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Both the mass spectrometric and the emission methods lend themselves to the use of very small samples, for example as little as 5 milligrams. This does not mean, however, that these instruments are the most sensitive for the detection of lowest concentrations of Deuterium Oxide in water. On the mass spectrometer or the emission spectrograph, for highest sensitivity, it is advisable when handling very small samples of water for analysis, to reduce the aqueous material to the Deuterium and Hydrogen gases. Dubbs²⁰ and Chinard and Ems²¹ have described methods for this purpose, using hot Zinc as the reducing agent. These papers would definitely have a bearing on the applications of any emission type instrument we might develop.

The mass spectrometric methods have disadvantages in that the instrumentation is expensive and for the most sensitive results the sample needs suitable preparatory procedures such as reduction to the elemental gases or the conversion to Methyl Deuteride and Methane. Spectrographic type instruments are cheaper but high sensitivity on low concentrations of Deuterium Oxide in water has not been proven as yet*. Here again conversion to the elemental gases seems desirable. Spectrographic equipment is also not widely distributed among those chemists who are most likely to require Deuterium Oxide analysis.

More sensitive methods seem desirable, methods which could either be applied to instruments more widely distributed among organic

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20. G. A. Dubbs, Anal. Chem. 25, 828 (1953).

21. F. P. Chinard and T. Ems, Anal. Chem. 25, 1413 (1953).

* According to further information received from Dr. J. L. Sanderson, this has been done by Broida and company at the National Bureau of Standards, but is unpublished to date.
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chemists, and biochemists, or the principles of which might lend themselves to the development of simpler and less costly instruments. Improved emission methods have been under consideration in this laboratory, and research undertaken. There remains a further possibility, and that is infrared absorption which heretofore has not been given serious consideration, either by us or by others.

In 1950 Thornton and Gondon²² described an infrared method of low sensitivity for the range 0 to 100% Deuterium Oxide in water. An accuracy of 3% was claimed, but it was expected that this could be improved upon. The method was based on the shift of the 2.5 micron water band to 3.8 microns in Deuterium Oxide. Tremner and Walker²³ made a more thorough investigation of the Thornton and Gondon method and discovered that the intensity of the 3.8 microns band for Deuterium Oxide was very sensitive to temperature changes. They developed a thermostated cell with which it was possible to analyze for Deuterium Oxide, with an accuracy of $\pm 0.1\%$ on 2% Deuterium. The Tremner paper is slightly vague in that the authors speak constantly in terms of Deuterium, though the analysis is based on heavy water. For heavy water, the above values of Deuterium should be multiplied by nine. This work was done on a single beam instrument and the analytical range considered was 0 to 3% Deuterium (0 to 45% Deuterium Oxide). These methods were not sensitive and the full potential sensitivity of the infrared technique such as might be obtained from a double beam instrument and differential methods had not been studied. These seemed worth investigating.

22. V. Thornton and E. F. Gondon, Anal. Chem. 22, 690 (1950).

23. H. R. Tremner and R. W. Walker, Instrument News, 4, No. 1, 1 (1952).

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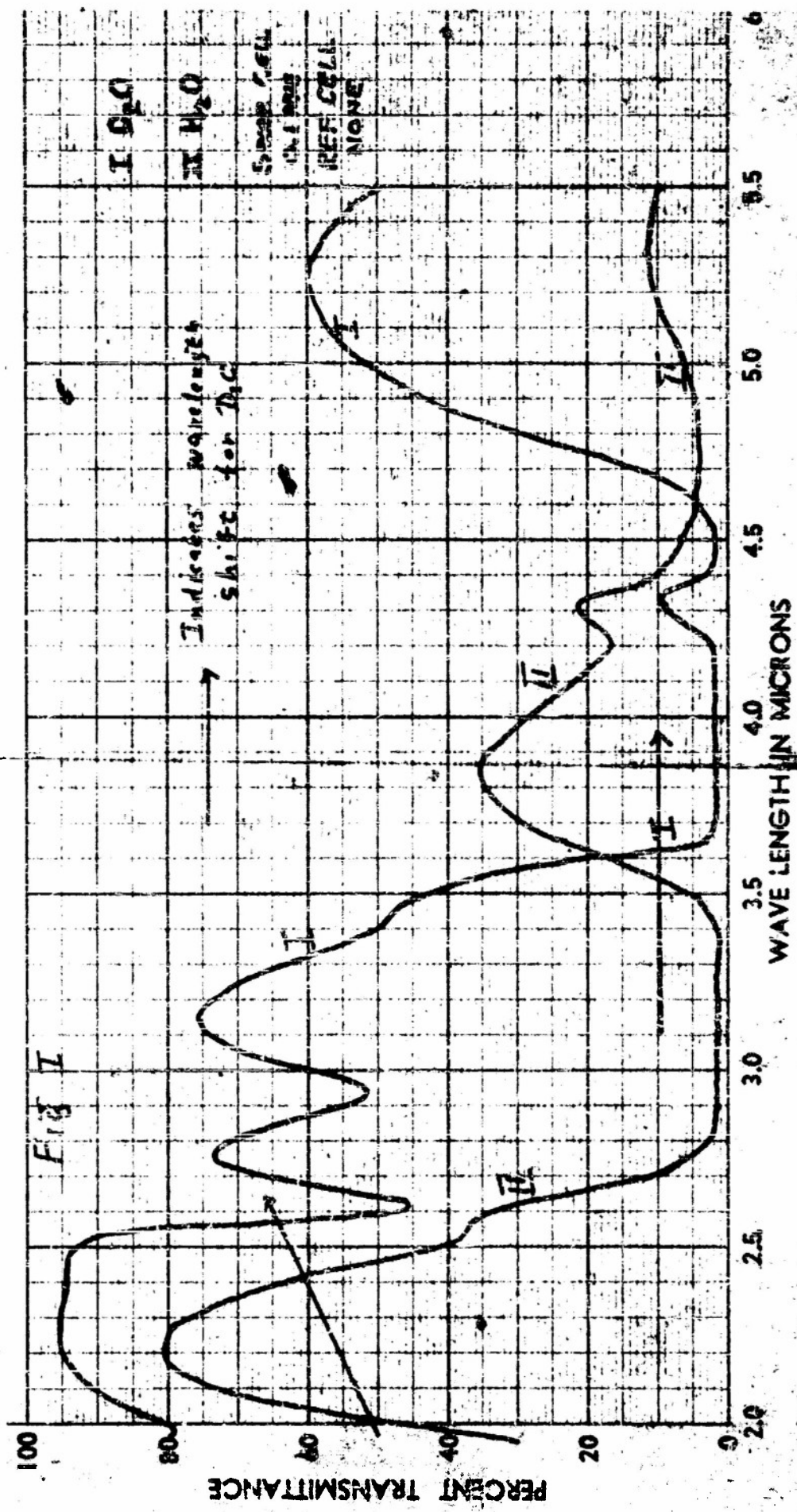
Object

The object of this investigation was to apply the most sensitive techniques of infrared spectroscopy to reach some conclusion on what are the lower limits of detection of Deuterium Oxide in the presence of water. In this initial work, precision and reproducibility were not considered. These would necessarily be the subject of another and more extensive investigation.

Experimental

1. The Use of 0.1 mm Cells

The most suitable thickness of an infrared absorption cell for water is something less than 0.01 mm depending on the problem. Trenner²² was able to use a thickness of 0.02 mm for the measurement of Deuterium Oxide. Any thicknesses greater than these would give spectra showing very high absorptions in most regions from 2 to 16 microns. Figure I shows the spectra of water and heavy water in 0.1 mm Calcium Fluoride cells with a Calcium Fluoride prism. The use of Calcium Fluoride cells is necessary because of the solubility of rock salt in water. However the use of the Calcium Fluoride prism in this particular instance had no significance. Studying the spectra, it is evident that the strongest absorption bands for water and Deuterium Oxide are at about 3.0 and 4.0 microns. The 4 micron Deuterium Oxide band appears at a gap in the absorption of the water spectrum and would be suitable as an analytical band which is in agreement with the choice of other workers on this analysis. The 2.9 micron band in the heavy water spectrum is evidently due to something less than 0.3% water as the Deuterium Oxide was only guaranteed to be at least 99.3%



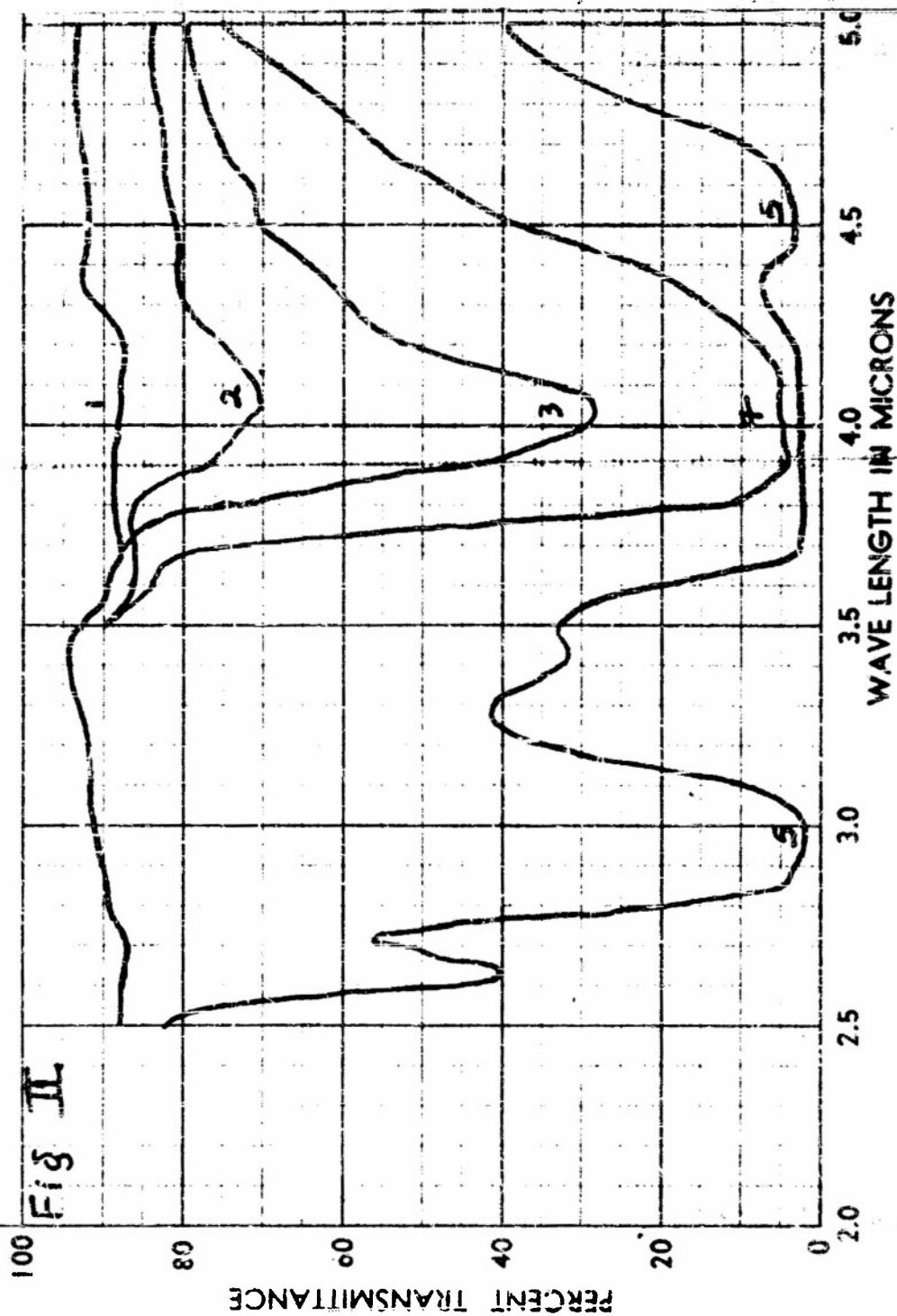
I. R. SPECTROPHOTOMETER

NO.		DATE		INDEX	
SAMPLE					
1. H_2O vs H_2O					
2. 0.15-0.2% D_2O in H_2O					
3. 1% D_2O in H_2O vs H_2O					
4. 10% D_2O in H_2O vs H_2O					
FROM S. APP. 3% H_2O in D_2O					
NO REF CELL					
SAMP. CELL		0.1 MM		CMS.	
REF. CELL		0.1 MM		CMS.	
CHEM.	MG.		MG.		
SOLV.					
VOL.	C.C.	%	C.C.	%	
F.S.	SOLID	C.	GAS	MM	

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Fig II

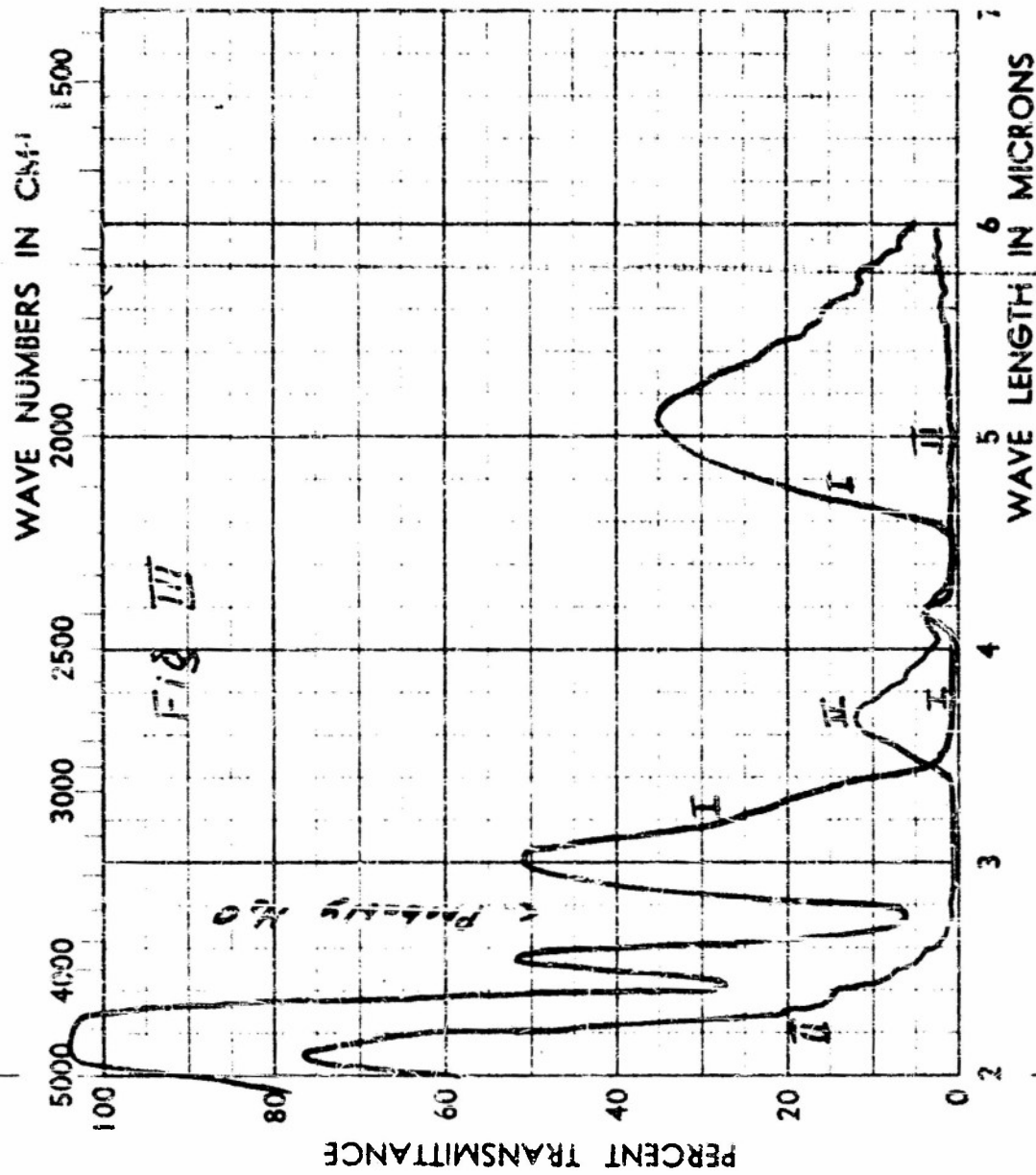


I. R. SPECTROPHOTOMETER
NaCl PRISM

NO.	DATE	INDEX
SAMPLE		
I D ₂ O		
II H ₂ O		
FROM		
SAMP. CELL=0.2MM	CM5.	
REF. CELL None	MM	CM5.
CHEM.	MG.	MG.
SOLV.		
VOL.	C.C.	%
F.S. SOLID	C.	GAS
		MM

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On Figure IV are shown the six standards run against distilled water in the reference beam, using matched 0.2 mm cells. These spectra were made using fixed slits, the width of which had been adjusted to give a D.C. voltage of 2.0 volts at the mechanical rectifier, at a wavelength setting of 3.7 microns, and with an 0.2 mm cell containing water in the sample beam and an opaque in the reference beam. Gains of voltage and Brown amplifier were as usual. This was also the procedure in adjusting the slit widths in subsequent work, although the actual voltage figure will vary.

Using as the base line the curve for H₂O against water, the absorbance values of these absorption bands were calculated. The results are plotted on Graph I. Observe the nice straight line relationship and the high degree of correlation between the different results; even when the 0.01 to 0.1% D₂O in H₂O range was expanded as shown in the right hand curve.

3. The Use of 0.5 mm Cells

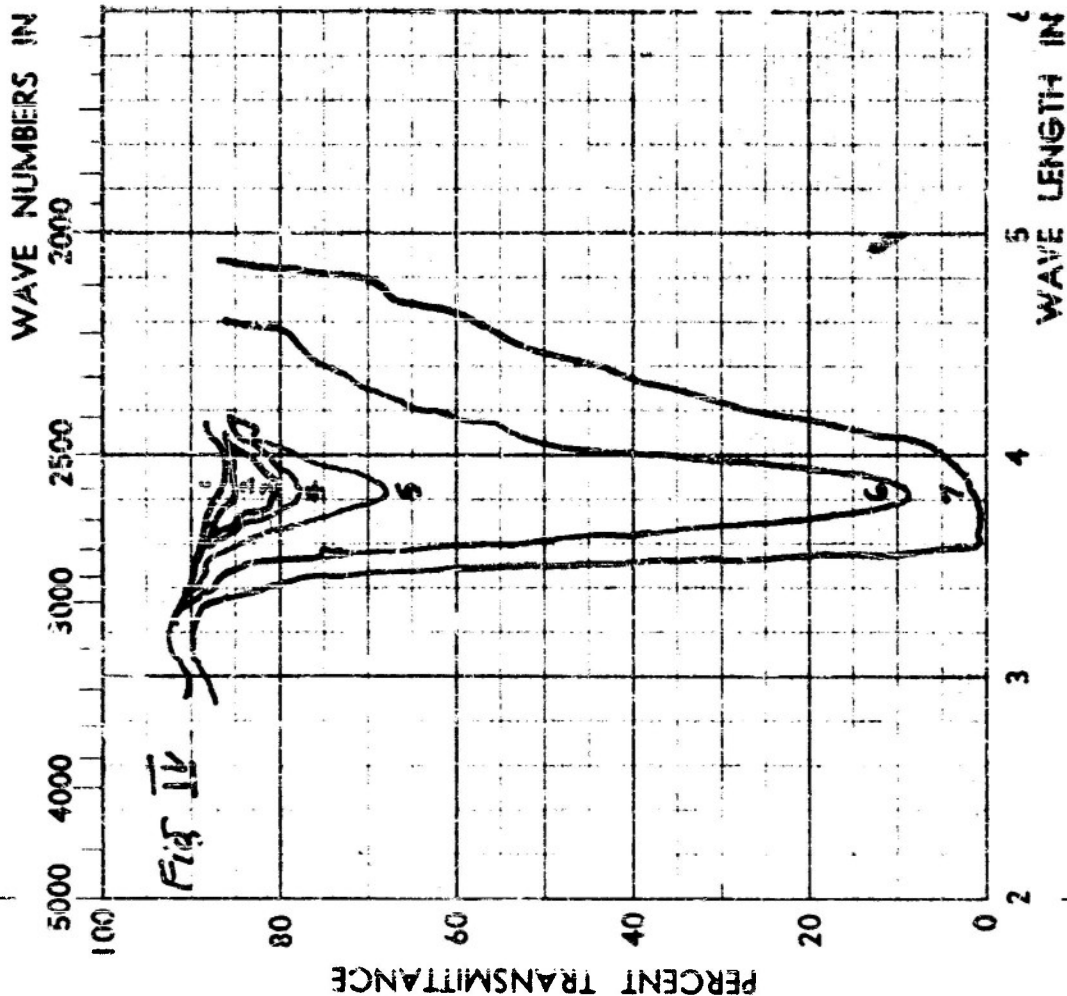
Since 0.5 mm cells instead of 0.2 mm cells should in theory give increased sensitivity, this was the logical next step. The spectrum of water in an 0.5 mm cell showed 99% absorption in the 3.8 to 4.0 micron region. This did not look very hopeful. Proceeding however to open up the slits to 0.5 volts as per our previous mentioned adjustment procedure, attempts were made to run water against water in the 0.5 mm cells. The very wide slits, however, introduced a complication in that much scattered light was introduced. This caused considerable trouble. For example, between 3.2 and 3.4 microns where there should be complete absorption in

I.R. SPECTROPHOTOMETER
NaCl PRISM

NO.	DATE	INDEX
SAMPLE		
1. H_2O vs H_2O		
2. 0.018% D_2O in H_2O vs H_2O		
3. 0.03% D_2O in H_2O vs H_2O		
4. 0.044% D_2O in H_2O vs H_2O		
5. 0.077% D_2O in H_2O vs H_2O		
6. 0.938% D_2O in H_2O vs H_2O		
FROM 7. 9.98% D_2O in H_2O vs H_2O		
SAMP. CELL= 0.2 MM		CMS.
REF. CELL 0.2 MM		CMS.
CHEM.	MG.	MG.
SOLV.		
VOL.	C.C.	% C.C.
F.S. SOLID	C.	GAS MM

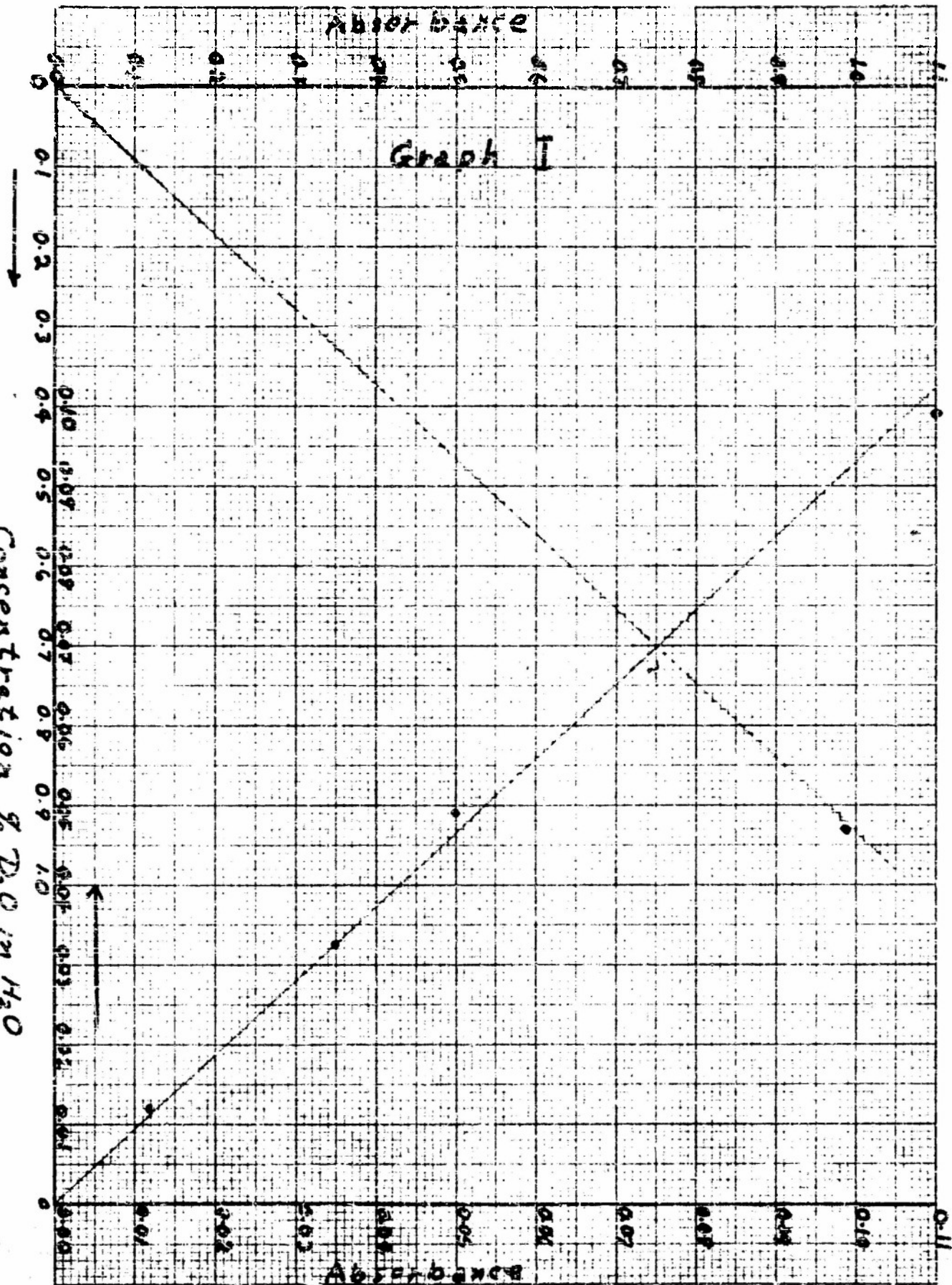
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Concentration % D_2O in H_2O



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both beams, there was a strong down scale drift, which could be reversed as the slits were narrowed, indicating definitely that there was an unbalance due to scattered light when the slits were set for the 0.5 volt reading. The instrument had a normal up scale drift. The down scale drift was so strong that it played havoc with all the curves and none was reproducible. It was not even possible to get a reproducible water vs. water baseline or reference to determine whether the two cells were matched, though of course this could be checked with other solvents. It should be noted that this down scale drift only occurred when there was complete absorption. Where absorption was not complete the pen then went up scale, trying to go back to its proper balance position. This meant that there was little chance for recording at wavelengths prior to the actual absorption band, but had to record immediately into the band or where there was energy, which in this case amounted to the same thing.

Considerable time was spent in studying this scattered light problem, full details of which were given in notebook 27400. A description of the tests carried out would only complicate this report, and besides it is an instrumental problem rather than an analytical one. Though it is the key to getting the highest sensitivity. All that need be said at this time is that some partial success was obtained even with slit settings of 0.5 volts at 3.7 microns. Much was still to be desired however, and it is quite probable that the sensitivity attained in this work could be at least doubled if a completely satisfactory answer to the scattered light problem were to be found.

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Figure V shows the first successful curve, run differentially, with Deuterium Oxide standards in the 0.5 mm cells. The absorbance values as shown in Graph 2 are only about 30% higher than those obtained using 0.2 mm cells. This increase in absorbance should have been about 2 1/2 times. The low values can be attributed almost entirely to the slantiness of the pen due to the low energy levels. It should be realized that the 0.5 volt setting was at 3.7 microns which meant that at about 3.9 to 4.0 microns the amount of energy would be on the order of 0.1 volt or less in both beams.

4. Using Reversed 0.5 mm Cells

Robinson²⁴ has shown that increased sensitivity can be obtained by first running samples differentially in the normal way and then recording again above the first spectrum but with the samples reversed in the two beams. This gives two curves which are mirror images of each other, the distance between the two peaks being an amplified absorbance value for the concentration in question. To do this properly only the samples are reversed, and not the cells, and the test then compensates for thickness discrepancies between the two cells. In the present work this refinement of technique was not deemed necessary and both cells and solutions were reversed.

The successful application of the Reversed Cell technique in this particular problem (only) required a reliable 100% line as starting point for recording both the upward and downward curves. This does not mean a true 100% line, but simply a starting point at which the pen could

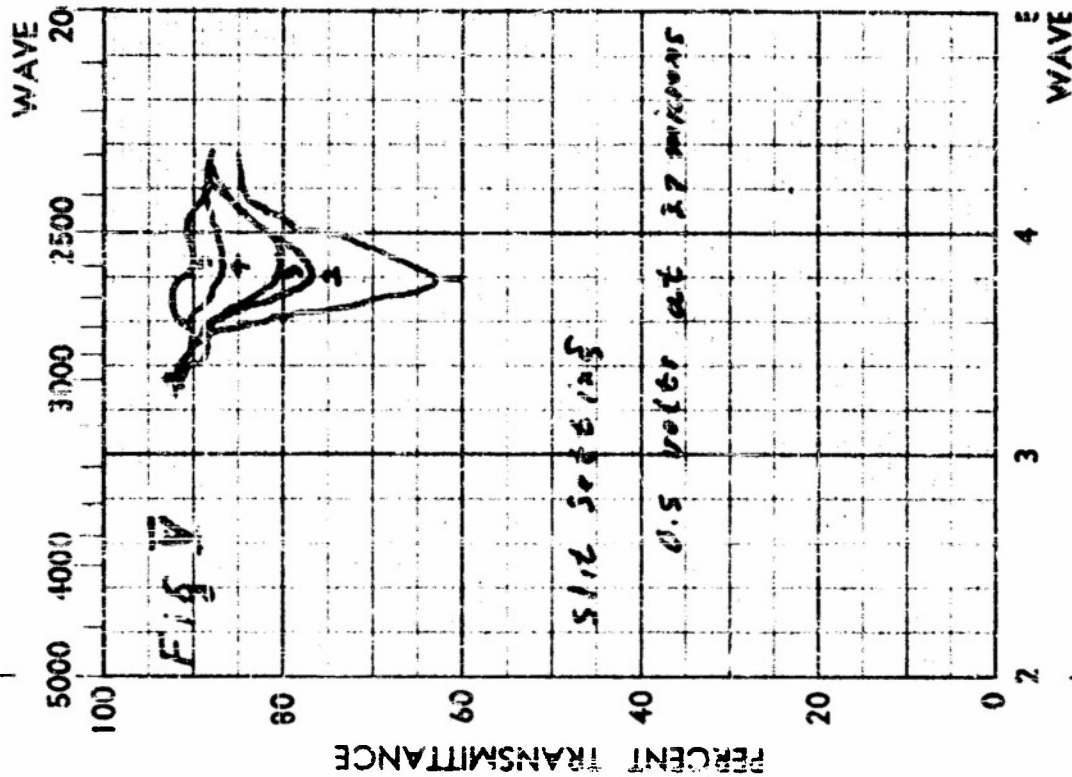
.....
24. D. Z. Robinson, Anal. Chem. 24, 519 (1952).
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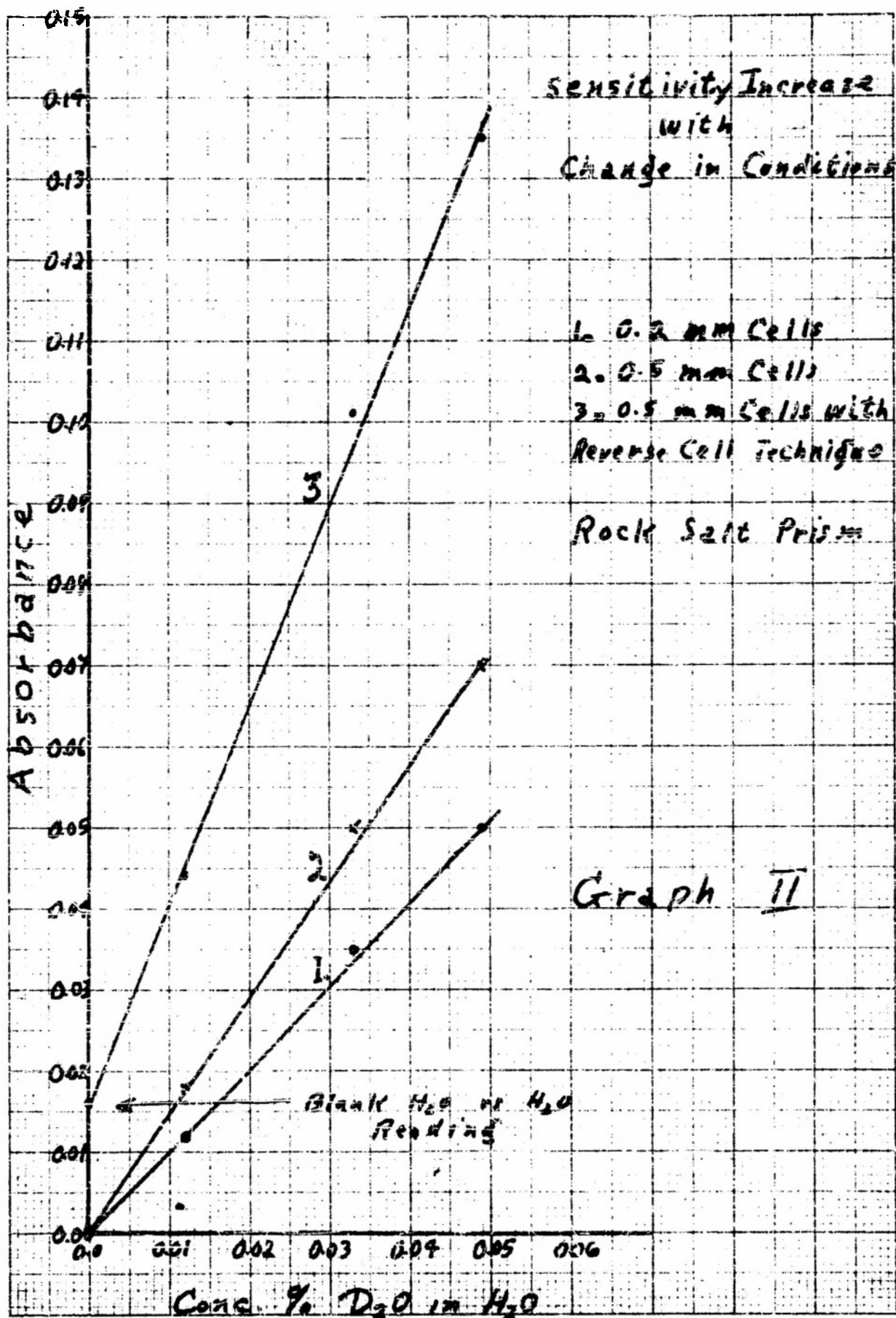
I. R. SPECTROPHOTOMETER
NaCl PRISM

NO.		DATE	INDEX
SAMPLE			
1.	0.099%	D ₂ O - H ₂ O	mm H ₂ O
2.	0.049%	D ₂ O - H ₂ O	mm H ₂ O
3.	0.033%	D ₂ O - H ₂ O	mm H ₂ O
4.	0.012%	D ₂ O - H ₂ O	mm H ₂ O
FROM 5. H ₂ O - H ₂ O			
SAMP. CELL = 0.5 MM		CMS.	
REF. CELL = 0.5 MM		CMS.	
CHEM.	MG.	MG.	
SOLV.			
VOL.	C.C.	%	C.C. %
F.S. SOLID	C.	GAS	MM

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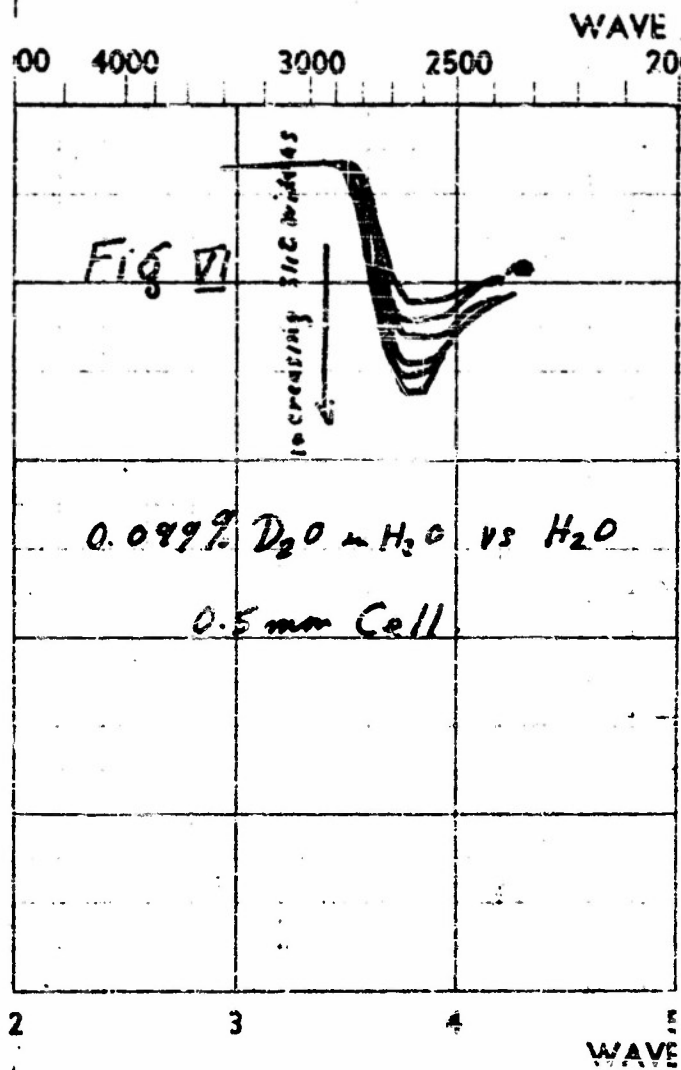


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be set and the position of which would not change during the recording until the scanning entered the D_2O absorption band region of the spectrum. This meant no down scale drift from scattered light. It also meant a low noise level. To attain the latter the voltage amplifier was switched to the 8x position and to obtain a responsive pen without scattered light interference the slits were gradually opened and curves recorded, noting the stability of the starting point. The series of curves obtained, using the 0.099% Deuterium Oxide standard, as the slits were widened are shown in Figure VI. No attempt was made to record the D.S. voltage on these slit openings. Using the slit opening designated by the lowest curve on Figure VI, four standards were recorded along with water with the reverse cell technique using 0.5 mm cells. The results are shown in Figure VII. The flat maxima of these curves is due to lack of sufficient energy between the two beams actuating the servo mechanism. Figure VIII shows the curve obtained from the same standard using A.S. bias in which the curves take their normal shape.

5. Discussion

On Graph 2 are plotted the absorbance values for the three standards 0.012, 0.053 and 0.049% D_2O in H_2O which were obtained during the course of this investigation as increasingly sensitive methods were applied. These absorbance values are from Figures IV, V and VIII. As mentioned previously the expected increase of 2 1/2 times the absorbance values on going from 0.2 to 0.5 mm thick cells did not occur. As also mentioned previously this is very probably due to the low energy levels involved which had to be kept low as wide slit openings introduced scattered light problems.

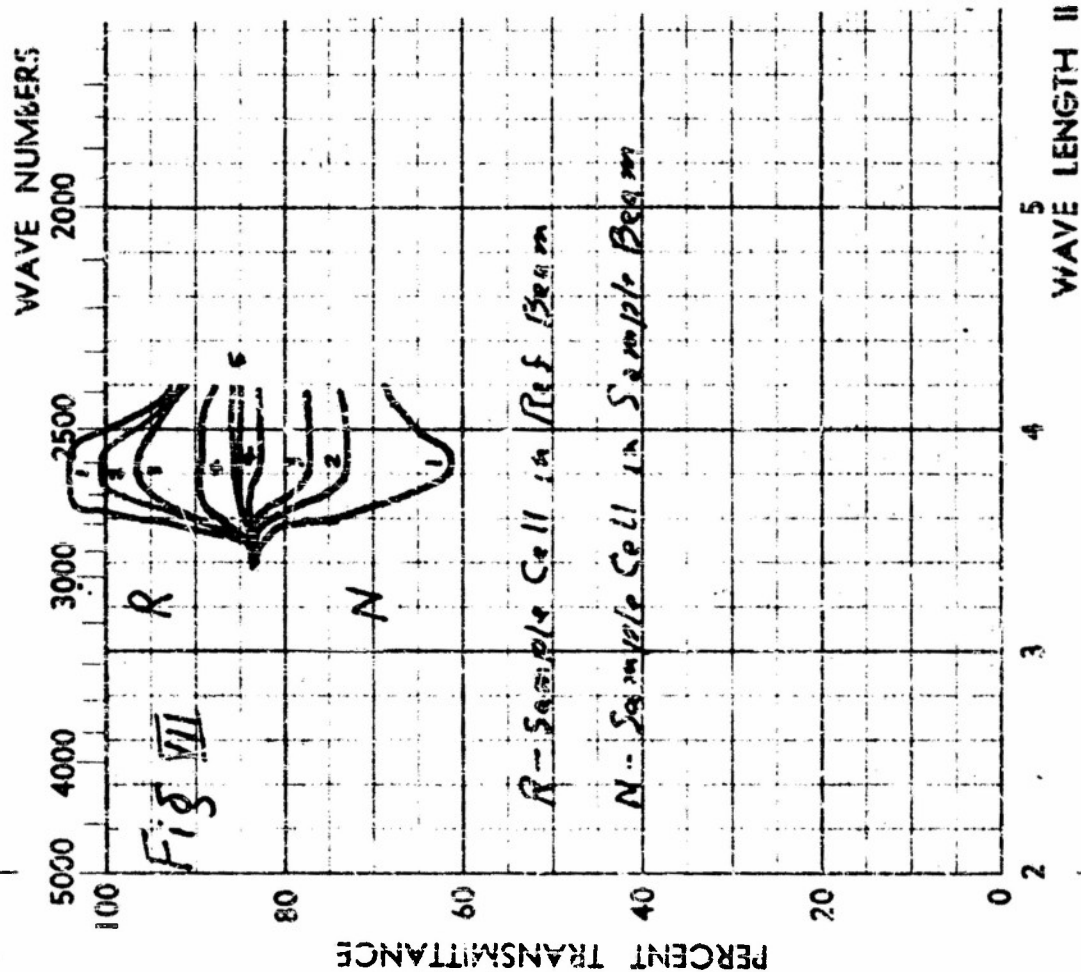


I. R. SPECTROPHOTOMETER
NaCl PRISM

NO.	DATE	INDEX
SAMPLE		
1.	0.099% D ₂ O in H ₂ O vs H ₂ O	
2.	0.049% D ₂ O in H ₂ O vs H ₂ O	
3.	0.073% D ₂ O in H ₂ O vs H ₂ O	
4.	0.022% D ₂ O in H ₂ O vs H ₂ O	
FROM 5 H ₂ O vs H ₂ O		
SAMP. CELL = 0.5 MM CMS.		
REF. CELL 0.5 MM CMS.		
CHEM.	MG.	MG.
SOLV.		
VOL	C.C.	% C.C. %
F.S. SOLID	C.	GAS MM

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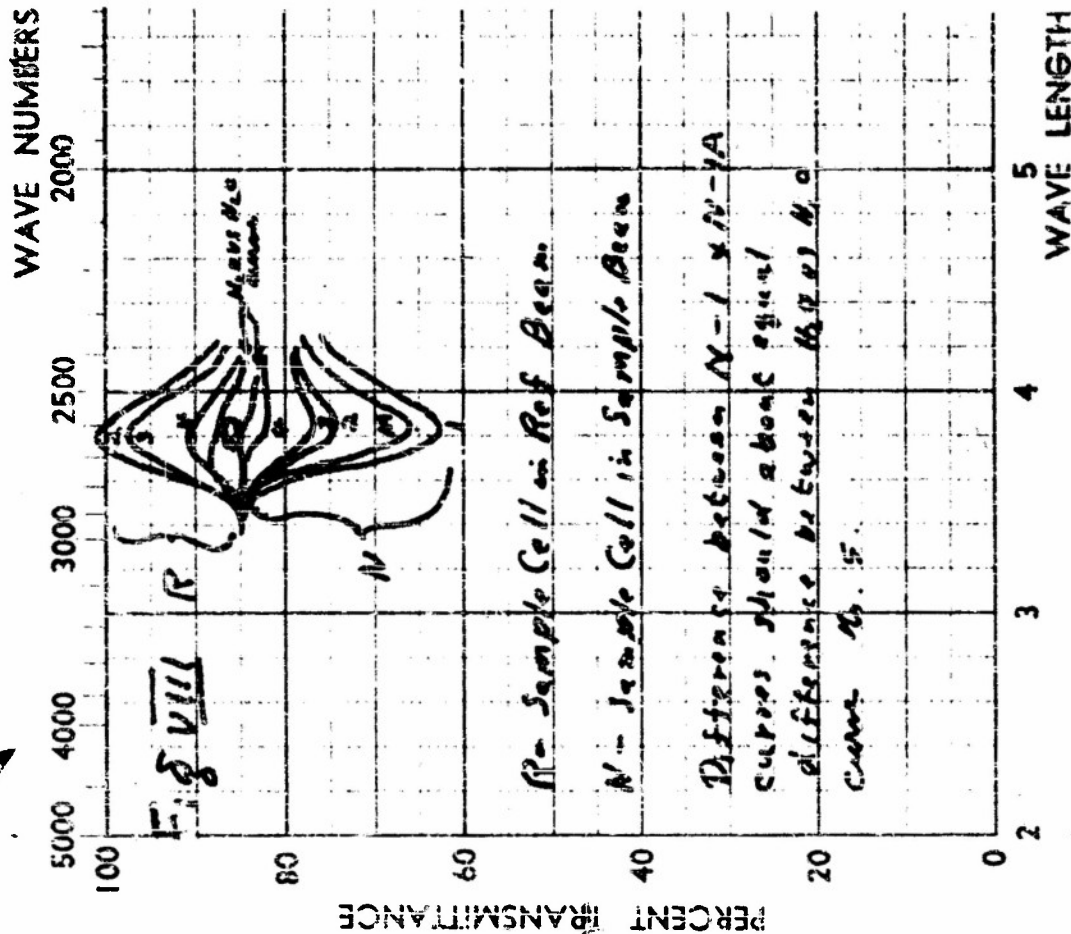


I. R. SPECTROPHOTOMETER NaCl PRISM

NO.	DATE	INDEX
SAMPLE		
1.	0.0088 g. H_2O vs H_2O	
2.	0.0458 g. D_2O vs H_2O	
3.	0.0378 g. H_2O vs H_2O	
4.	0.0128 g. H_2O vs H_2O	
5.	H_2O vs H_2O	
FROM 1A. 0.0378 g. D_2O vs H_2O		
REF. CELL: 6.5 MM		
REF. CELL: 6.5 MM		
CHEM.	MG.	MG.
SOLV.		
VOL.	C.C.	% C.C. %
F.S. SOLID	W.C.	GAS MM

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On passing to curve 3 on Graph 2, that for the reversed cell technique, the results are completely in line, being about twice those obtained for the standard method using 0.5 mm cells (curve 2). Note that curve 3 does not go to zero but to 0.015 absorbance value which is the absorbance difference between the two cells filled with water as indicated in Figure VIII. If solutions had been reversed but not the cells, this curve 3 of Graph 2 should have gone to zero, as with curves 1 and 2. A glance at the absorbance bands of Figure VIII and at curve 3, Graph 2 makes it readily apparent that 0.01% Deuterium Oxide added to ordinary water is readily measurable. It is also apparent that concentrations as low as 0.005% Deuterium Oxide could be detected without too much difficulty. The correlation between the points on curve 3, Graph 2 indicates that the percent Deuterium Oxide in the range 0.01 to 0.05% could probably be determined with an accuracy of $\pm 0.001\%$ if a precisely controlled method were developed.

Conclusions

While final conclusions should not be drawn from a single series of experiments, there seems little doubt that the infrared absorption method is fully capable of detecting and measuring 0.01% Deuterium Oxide in water. The possible degree of precision in the analysis could be about $\pm 0.001\%$ or better.

Work so far has indicated that percentages of Deuterium Oxide as low as 0.005% are within the sensitivity limit of this method. The ultimate sensitivity of the infrared method has not been reached in this work due to restrictions on the limits of the experimental technique from scattered light. If these were removed a sensitivity of 0.001% does not seem out of the way.

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APPENDIX II

Bibliography

2.1 - 2.8 Previous Technical Reports Submitted by Baird Associates, Inc.
under Contract No. NSCmr-80601

3.1 - 3.16 Published Papers and Talks. (See detailed list and abstracts,
Section I of this report.)

Footnotes:

1. B. H. Billings, J. Opt. Soc. Am. 27, 738 (1947).
2. B. Lyot, *Script. rend.* 197, 1593 (1953).
3. W. P. Mason, *Phys. Rev.* 62, 173 (1946).
4. Zwicker and Scherrer, *Helv. Phys. Acta* 17, 546 (1944).
5. R. O'E. Carpenter, J. Opt. Soc. Am. 40, 223 (1950).
6. B. Lyot, *Script. rend.* 197, 1593 (1953); Y. Ohnuma, *Nature* 141, 291 (1958).
7. B. H. Billings, J. Opt. Soc. Am. 27, 738 (1947).
8. G. Dufour, Thesis, Paris (1950).
9. P. Jacquinet and G. Dufour, *J. Recherches du C.N.R.S., Bellevue*, 6, 1 (1948).
10. H. D. Polster, J. Opt. Soc. Am. 39, 1038 (1949).
11. Mohls, *Ann. d. Phys.* 22, 433 (1957).
12. Pierre Giaccone, *Script. rend.*, December 1952, p. 1637.
13. William S. Fastie, J. Opt. Soc. Am. 42, 641, 647 (1952).
14. John W. Evans, J. Opt. Soc. Am. 39, 229 (1949).
15. B. W. Thomas, *Anal. Chem.* 22, 1476 (1950).
16. O. Orchin, I. Wender and R. A. Friedel, *Anal. Chem.* 21, 1672 (1949).
17. A. S. Keston, D. Rittenberg and R. J. Schoenheimer, *J. Bio Chem.* 122, 227 (1957).

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18. M. P. Broida and J. W. Meyer, J. Opt. Soc. Am. 48, 37 (1958).
19. H. J. Morawitz and M. P. Broida, Anal. Chem. 34, 1577 (1962).
20. G. A. Dubbs, Anal. Chem. 25, 828 (1953).
21. F. P. Chinnard and T. Ems, Anal. Chem. 25, 1413 (1953).
22. V. Thornton and E. R. Gordon, Anal. Chem. 22, 690 (1950).
23. H. R. Trenner and R. W. Walker, Instrument News, 4, 1 (1952).
24. D. L. Robinson, Anal. Chem. 34, 619 (1962).

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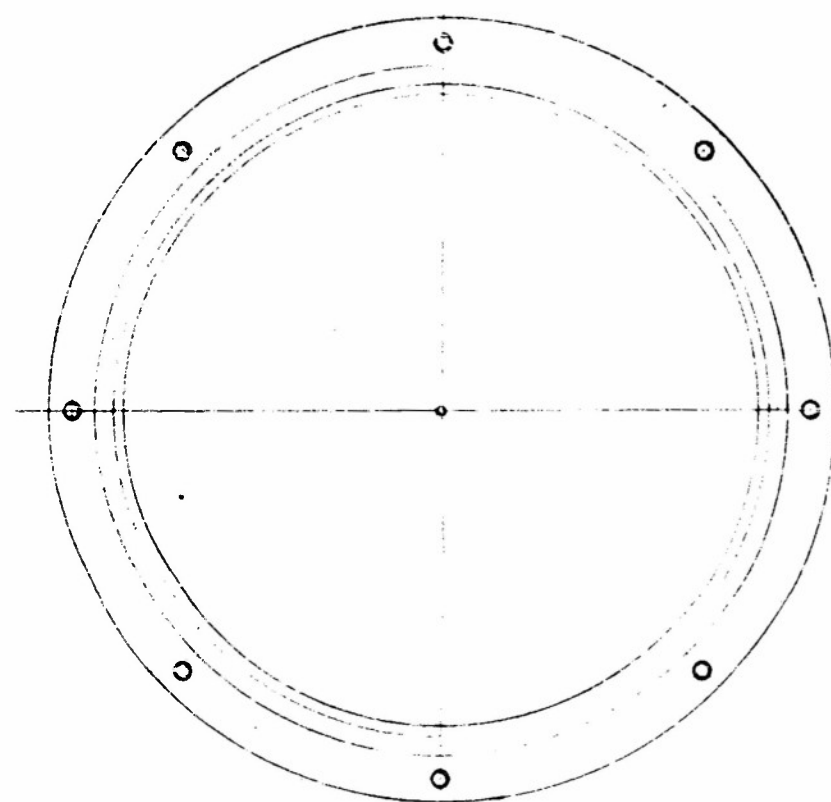
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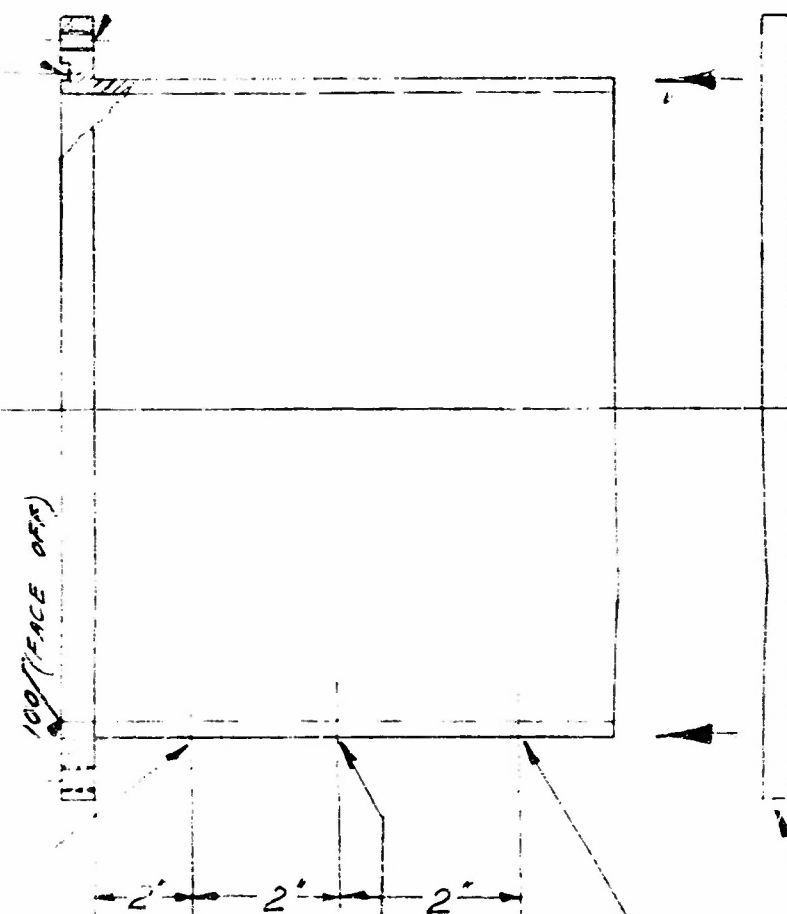
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